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Paleoecological Analysis of Lake Acidification Trends in North America and Europe Using Diatoms and Chrysophytes

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16. ABSTRACT

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Paleoecological studies indicate that recent acidification has been caused by acidic deposition in the Adirondack Mountains (New York), northern New England, Ontario, Quebec, and Canadian Atlantic provinces in North America; England, Scotland, and Wales in the United Kingdom; Norway, Sweden, Finland, The Netherlands, and West Germany in Europe. Inferred pH decreases are commonly as much as 0.5-1.5 pH units. No acidification trends were observed in regions currently receiving low deposition of strong acids (e.g., Rocky Mountains and Sierra Nevada in the western United States, northwest Norway, and northwest Scotland). Slight or no trends towards decreasing pH were observed in study lakes receiving moderately acidic deposition (Upper Midwest and northern Florida, United States). The magnitude of pH decline in lakes studies is greater, on the average, in Europe than in North America. The amount of inferred acidification (increase in H^+ concentration) correlates with the amount of S and N loading and the ability of watersheds and lakes to neutralize acid inputs, and is generally consistent with current lake acidification models.

In most cases, the primary cause of recent acidification trends (post-1850) is atmospheric deposition of acidic material, as opposed to land use changes or natural processes, though these may be contributing factors. Acid loading has decreased in some regions since 1970 (e.g., northeastern United States; United Kingdom). Some lakes have become less acidic in response, but others continue to lose buffering capacity and are becoming more acidic. Many currently acidic lakes were naturally acidic (pH < 5.5) prior to the onset of anthropogenic acidification. These lakes are typically small (< 10 ha), located at moderately high elevations, have thin or peaty soils, or are located in outwash deposits. Many of these have acidified further recently.

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PALEOECOLOGICAL ANALYSIS OF LAKE ACIDIFICATION TRENDS IN NORTH AMERICA AND EUROPE USING DIATOMS AND CHRYSOPHYTES

Final Draft (Fourth)

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Abstract

Analysis of sediment diatom and chrysophyte assemblages is the best technique currently available for inferring past lakewater pH trends, and use of this approach is increasing rapidly. Sediment core inferred pH data exist for at least 100 lakes in both North America and Europe. This number will approximately double within the next two years. The pH inference equations are based on at least 15 calibration data sets for North America and 10 for Europe, involving totals of at least 500 and 300 lakes, respectively.

Paleoecological studies indicate that recent acidification has been caused by acidic deposition in the Adirondack Mountains (New York), northern New England, Ontario, Quebec, and Canadian Atlantic provinces in North America; England, Scotland, and Wales in the United Kingdom; Norway, Sweden, Finland, The Netherlands, and West Germany in Europe. Inferred pH decreases are commonly as much as 0.5-1.5 pH units. No acidification trends were observed in regions currently receiving low deposition of strong acids (e.g., Rocky Mountains and Sierra Nevada in the western United States, northwest Norway, and northwest Scotland). Slight or no trends towards decreasing pH were observed in study lakes receiving moderately acidic deposition (Upper Midwest and northern Florida, United States). The magnitude of pH decline in lakes studied is greater, on the average, in Europe than in North America. The amount of inferred acidification (increase in H^+ concentration) correlates with the amount of S and N loading and the ability of watersheds and lakes to neutralize acid inputs, and is generally consistent with current lake acidification models.

In most cases, the primary cause of recent acidification trends (post-1850) is atmospheric deposition of acidic material, as opposed to land use changes or natural processes, though these may be contributing factors. Acid loading has decreased in some regions since 1970 (e.g., northeastern United States; United Kingdom). Some lakes have become less acidic in response, but others continue to lose buffering capacity and are becoming more acidic. Many currently acidic lakes were naturally acidic (pH < 5.5) prior to the onset of anthropogenic acidification. These lakes are typically small (< 10 ha), located at moderately high elevations, have thin or peaty soils, or are located in outwash deposits. Many of these have acidified further recently.

Additional Index Words: Paleolimnology, sediment cores, pH, alkalinity, historical trends

I. Introduction

Acidification of lakes by atmospheric deposition of strong acids is a major environmental issue and causes and effects have been studied intensively. However, there are still major uncertainties concerning the acidity status of lakes prior to the onset of acidic deposition, and how these lakes have changed in response to inputs of acids. More information on these topics is necessary to fully assess the effects on aquatic resources and to accurately predict the nature and extent of response to future changes in atmospheric deposition. Historical changes have been assessed by (1) comparison of recent and historical water chemistry, fish, and diatom data, (2) use of empirical and dynamic models, and (3) analysis of paleoecological data (National Research Council, 1986; Davis and Stokes, 1986). Of these, paleoecological studies have provided some of the best information. Other than the sparse chemistry and fisheries information, which is usually difficult to interpret, it provides the most direct indication of trends in acidity status. Also, new paleoecological studies of lakes can be made at any time. They do not depend on the existence of historical records. Paleolimnological studies can provide information on past lake pH and acid neutralizing capacity (ANC) and how it has changed through time, the rate and magnitude of the change, possible causes of the change, and the relative importance of processes and mechanisms (Battarbee, 1984; Charles & Norton, 1986; Smol et al., 1986).

Because of the usefulness and importance of information on lake acidification that can be derived from paleoecological investigations, there has been a rapid increase in the number of such studies in Europe and North America (Battarbee & Charles, 1986, 1987). This chapter provides an up-to-date assessment of the studies, first by describing the underlying rationale, assumptions, and methods, and then by summarizing, comparing, and contrasting the results of studies on the two continents.

Much of the background information on method and approach is derived from Charles and Norton (1986). Summary and assessment of studies in the different geographic regions are treated by the authors as follows: Charles, United States; Smol, Eastern Canada; Battarbee, United

Kingdom; Renberg, Norway, Sweden, and Finland; and Van Dam, Austria, Belgium, Czechoslovakia, Denmark, The Netherlands, and West Germany.

The sediments of a lake contain information on the lake's past: its biota, water chemistry, and watershed characteristics, and material it has received directly from the atmosphere (Frey, 1969; Pennington, 1981). The information is provided by the organic and inorganic substances, in dissolved or particulate form, that entered or were formed within a lake and deposited in its sediments. The primary materials are controlled by watershed geology, climate, and biological processes. Once deposited at the bottom of a lake, sediments can be affected by secondary processes, such as transport (horizontal and vertical) and a variety of chemical and biological activities.

Sediments can be sampled by taking cores; the core samples represent the history of sediment deposition, with the older sediments lying at greater depth. The times when particular intervals of sediment were deposited can be determined either by analysing the radioactive content (^{210}Pb, ^{137}Cs, ^{14}C) or by examining changes in sediment characteristics, such as pollen and charcoal as indicators of logging and forest fires, that correspond to well-dated local events.

Several components of sediments provide information on factors related to lake acidification. Concentrations of polycyclic aromatic hydrocarbons (PAH), sphaerical carbonaceous particles (soot), lead, sulfur, vanadium, sulfur isotope ratios, and magnetic particles can be interpreted to indicate trends in atmospheric deposition of substances derived from combustion of fossil fuels. The pH of lake water during the past can be inferred by analyzing assemblages of diatoms and chrysophytes. Remains of chydorids (littoral crustaceans) and chironomids (midge larvae) may also provide insight into changes in aquatic biota related to acidification.

II. Inferring Past Lakewater pH: Rationale and Methods

Analyzing and interpreting sediment diatom and chrysophyte assemblages is the best paleolimnological technique available for inferring past lakewater pH. Investigators are using this approach increasingly to assess changes that may have been caused by atmospheric deposition of

strong acids, because patterns of change in lakewater pH can be used to help determine trends in acidic deposition.

A. Diatoms and Chrysophytes as Indicators of Lake Chemistry

Diatoms make up a large group of single-celled freshwater and marine algae (class Bacillariophyceae). They have siliceous cell walls and are formed of two halves or valves. Chrysophytes (Chrysophyceae) are primarily planktonic. In this chapter, the term chrysophyte refers to only one family, the Mallomonadaceae, also known as the scaled chrysophytes. Its members have flagella and an external cell covering of overlapping siliceous scales and bristles. The scales are used for paleoecological reconstructions.

The distributions of diatom taxa are closely related to water chemistry (Cleve, 1891; Kolbe, 1932; Hustedt, 1927, 1939; Jørgensen, 1948; Cholnoky, 1968; Patrick and Reimer, 1966, 1975; Patrick, 1977). For this reason, diatoms are commonly used as indicators of pH, nutrient status, salinity, and other water quality characteristics (e.g., Lowe, 1974). Stratigraphic analysis of fossil diatom assemblages can be used to investigate changes in lakes resulting, for example, from shifts in climate, development of watershed soils and vegetation, local human disturbance of watersheds, and acid deposition (Battarbee, 1984, 1986; Fritz and Carlson, 1982; Brugam, 1983).

Diatom assemblages in sediment are good indicators of past lake pH because (1) diatoms are common in nearly all freshwater habitats, (2) distributions of diatom taxa are strongly correlated with lake water pH (Hustedt, 1927, 1939; Merilăinen, 1967; Battarbee, 1984; Gasse & Tekaia, 1983; Gasse, 1986; Huttunen & Merilăinen, 1983; Dixit, 1983; Davis & Anderson, 1985; Charles, 1985; Anderson et al., 1986; Dixit, 1986b; Scruton & Elner, 1986; Flower, 1987; Taylor, 1986), (3) diatom remains are preserved well in sediment and can usually be identified to the lowest taxonomic level, (4) their remains are usually abundant in sediment (10^4 to 10^8 valves/cm^3 of sediment) so that rigorous statistical analyses are possible, and (5) many taxa are usually represented in sediment assemblages (20 to 100 taxa per count of 500 valves is typical), so that inferences are based on the ecological characteristics of many taxa.

Some disadvantages in using diatoms as pH indicators are that (1) diatom identification requires considerable taxonomic expertise, (2) diatoms are not well preserved in some cores because of dissolution (in some peaty and some calcareous sediments), (3) sometimes the number of taxa is low (e.g., in some peatland pools), (4) calibration data sets (the current relationship between water chemistry and surface sediment diatom assemblages) are not always available for the lake region studied, and (5) good ecological data are not always available for all dominant taxa. Other problems associated with interpretation of diatom data are discussed in Charles & Norton (1986), Battarbee (1986), and Davis & Smol (1986). Except for the dissolution problem, all the above disadvantages are equal to or less than disadvantages for other groups of aquatic organisms.

In general, the use of chrysophyte scales for pH reconstructions involves the same advantages and disadvantages as those for diatoms (Smol, 1980, 1987, 1988; Smol et al., 1984a,b; Kristiansen, 1986; Steinberg & Hartmann, 1986; Hartmann & Steinberg, 1987), except that the number of chrysophyte taxa in a sediment assemblage is typically one-tenth the number of diatom taxa and most chrysophyte taxa are euplanktonic (normally suspended in the water). The latter characteristic provides an advantage over diatoms in the study of acidic lakes because euplanktonic diatoms are usually rare or nonexistent in lakes with pH below about 5.5 to 5.8 (Battarbee, 1984; Charles, 1985; Renberg, et al., 1985). In these cases, chrysophytes may be more sensitive indicators of water chemistry changes than diatoms because they live in direct contact with the open water. Most diatoms grow in the shallower water of the littoral zone, which may be chemically different from the open water.

B. Techniques for Inferring Past pH

Several techniques based on diatom assemblages have been used to assess trends in acidification and to derive equations for inferring lakewater pH. These are described briefly below. Further descriptions and discussion of details and uncertainties associated with the reconstruction of lakewater pH are covered in depth by Gasse & Tekaia (1983), Battarbee (1984), Davis & Anderson

(1985), Charles (1985), Smol et al. (1986), Oehlert (1984), Taylor et al. (1986), Birks (1987), Ter Braak & Van Dam (in press), and Stevenson et al. (1988).

The simplest and most straightforward approach is to count sediment core diatom and chrysophyte assemblages and prepare depth profiles of percentages of the dominant taxa. Changes in the profiles are then interpreted in light of the ecological data available for those taxa. Until very recently, this was the only technique used to analyze chrysophyte scale data. Now, however, quantitative techniques have been developed (Charles & Smol, 1988; Dixit et al., 1988a).

Hustedt (1939; see also Battarbee et al., 1986) made one of the first significant steps toward establishing a more quantitative approach for using diatoms as pH indicators. He recognized the strong relationship between diatom distributions and lakewater pH and defined the following pH occurrence categories (from Battarbee et al., 1986):

1. Alkalibiontic: occuring at pH values > 7;
2. Alkaliphilous: occurring at pH about 7 with widest distribution at pH > 7;
3. Indifferent: equal occurrence on both sides of pH 7;
4. Acidophilous: occurring at pH about 7 with widest distribution at pH < 7;
5. Acidobiontic: occurring at pH values < 7 with optimum distribution at pH 5.5 and under.

Assignments of diatom taxa to these categories can be based on literature references and on the distribution of taxa within waters of particular geographic regions. Changes in the percentages of diatom valves in each pH category in a sediment core can be used to estimate trends in lakewater pH. This method makes use of data on most of the taxa within a core, not just the most common.

Nygaard (1956) took the next major step with the development of a set of indices. These indices are based on ratios of the percentages of diatom valves in Hustedt's pH categories. Acid units and alkaline units are calculated as an intermediate step:

acid units = 5 (% acidobiontic) + (% acidophilous),

alkaline units = 5 (% alkalibiontic) + (% alkaliphilous)

The percentages of valves in the two extreme pH categories, acidobiontic and alkalibiontic, are weighted by a factor of 5 because diatoms in these categories are presumably stronger indicators of pH.

The formulae for Nygaard's indices are:

$$\alpha = \frac{\text{acid units}}{\text{alkaline units}}$$

$$= \frac{\text{acid units}}{\text{number of acid taxa}}$$

$$\epsilon = \frac{\text{alkaline units}}{\text{number of alkaline taxa}}$$

Of these, index α is the best predictor of current pH in acidic lakes (Merilăinen, 1967; Renberg, 1976; Davis & Anderson, 1985; Charles, 1985; Beeson, 1984) and has been used most frequently. Application of index α to acidic lakes is limited, however, if diatom assemblages have no alkaliphilous or alkalibiontic taxa. In these cases there are zero alkaline units and the denominator of the index is zero. It is possible to circumvent this problem by arbitrarily setting the denominator equal to one (e.g., Charles, 1985) or some lower number. The index may still work reasonably well.

Merilăinen (1967) refined quantitative techniques even further by developing an approach for predicting lakewater pH from the index values. The relationship between the $\log_{10}$ of index

values and measurements of lakewater pH for several lakes is determined by using simple regression analysis. Predictive equations are then derived directly from the slope and intercept of the regression equations.

Renberg and Hellberg (1982) derived a new index (Index B), also based on pH categories, that can be used to quantitatively infer pH:

$$\text{Index B} = \frac{(\%\ \text{indifferent}) + 5(\%\ \text{acidophilous}) + 40\ (\%\ \text{acidobiontic})}{(\%\ \text{indifferent}) + 3.5\ (\%\ \text{alkaliphilous}) + 108\ (\%\ \text{alkalibiontic})}.$$

Index B has advantages over index α, including the use of more information and less reliance on alkaline taxa, which are typically rare or absent in acid lakes. Other indices incorporating Hustedt's (1939) pH categories have been developed (e.g., Watanabe and Yasuda, 1982).

Predictive equations can also be developed from multiple linear regression analysis of measured lakewater pH with the percentages of diatoms in each pH category (e.g. Davis & Anderson, 1985; Charles, 1985; Figure 1). Standard errors and confidence intervals for prediction of new points are usually smaller than those for the indices discussed above (e.g., Charles, 1985; Huttunen & Merilăinen, 1986b). Other multiple regression approaches for inferring pH involve the use of selected taxa or principal components of taxa data sets as independent variables (Davis & Anderson, 1985; Gasse & Tekaia, 1983). The standard error for inferred pH ranges between 0.25 and 0.5 pH units (Battarbee, 1984; Charles & Norton, 1986). Ter Braak & Van Dam (in press) have developed new methods for inferring pH and other chemical characteristics from diatoms using maximum likelihood calibration based on Gaussian logit response curves of taxa against pH, and weighted averaging. Oksanen et al. (1988) used weighted averaging, least squares, and maximum liklihood to calculate pH optima and tolerance of diatom taxa, and then used these estimates to predict the pH of other lakes, using weighted averaging. Detrended correspondence analysis (DCA) has been used to infer pH trends (Huttunen & Merilăinen, 1986). A promising new technique incorporating DCA and canonical correspondence analysis (CANOCO) (Ter Braak, 1986) can also be used to infer pH and other water chemistry characteristics as well (Stevenson,

et al., 1988). A multiple regression technique, similar to that for pH, has been developed to infer alkalinity (Charles et al., 1989). Also, there are new techniques to infer pH using both diatoms and chrysophytes (Charles & Smol, 1988). These are particularly useful in acidic lakes (pH ≤ 5.0), because chrysophytes appear more sensitive to pH changes in these ranges than do diatoms (e.g., Gibson et al., 1987).

Trends in pH curves usually are not analyzed statistically. Instead, subjective interpretations are made that account for the nature of the diatom assemblages, the error associated with the predictive techniques, the evidence of sediment mixing, and other factors. This is not a problem if pH changes are great, but relatively small changes (e.g., 0.2 - 00.3 pH units) must be interpreted cautiously, especially if the changes do not show a consistent trend over several sediment depth intervals. Esterby and El-Shaarawi (1981a,b) have developed a point-of-change technique to determine the point of maximum rate of change in a profile of dominant diatom taxa and whether the change is statistically significant. This technique has been used to evaluate taxa profiles from at least one lake (Delorme et al., 1983). The technique has also been applied to pH profiles (R. Kreis, pers. comm.). A Baysian statistical approach has been developed and used to assess different sources of uncertainty in calibration data sets and inference equations, but since it incorporates Monte Carlo simulation, it is too computer intensive to use for regular analysis of cores (Oehlert, 1984; in press).

Diatom and chrysophyte data can be used to address questions such as: Has a lake become more acidic or alkaline? How great were the changes? When did they occur? What were the causes? The extent to which these questions can be answered depends on (1) the quality of sediment cores, (2) the preservation and diversity of sediment diatoms, (3) the quality of diatom slide preparation and counting methods, (4) the quality of taxonomic identifications, (5) the precision and accuracy of the pH inference techniques and the applicability of the equations to a study region or lakes, (6) the accuracy and precision of dating and other information on sediment characteristics, (7) the availability of historical watershed and atmospheric deposition data, and (8) research design.

C Evaluation of Lake Acidification Causes

Diatom and chrysophyte data can be used not only to infer the past pH trend of a lake but also, in many cases, to suggest the causes of the changes. There are three major potential causes of acidification of low-alkalinity lakes: (1) long-term natural acidification, (2) watershed disturbances, such as fires, blowdown, logging, complete deforestation, reforestation and decrease in agricultural use of land (primarily in Europe), and ensuing responses of vegetation and soils, and (3) atmospheric deposition of strong acids. Other factors may affect lake acidification, but not normally on a regional scale. These include nearby emission sources, discharge of factory effluent, cultural development (roads and houses, for example), land clearance for agriculture, acid mine drainage, mixing with seawater, liming, paludification, drainage of wetlands, and water level changes such as those resulting from small dams, beaver activity, or changes in climate. The last factor is probably important only when a significant proportion of a watershed is wetland and net sulfur reduction-oxidation and cation exchange processes are affected. In shallow pools (e.g., in The Netherlands), low pH values may occur during dry summers because of oxidation of sulphides in the exposed sediments. Because these other factors have not, with few exceptions, affected the lakes evaluated in this report, they are not considered further.

The three primary potential causes of lake acidification are addressed below. Diatom studies of long-term acidification in both Europe and North America are also briefly reviewed. Following this is a summary of patterns of diatom changes to be expected in response to each major acidification cause.

1. Long-term Natural Acidification

Long-term trends in the postglacial development of Temperate Zone European and North American lakes have been studied using sediment diatom, chrysophyte, pollen, and chemical analyses. These studies indicate that most acidic to weakly alkaline lakes in areas having bedrock that is relatively resistant to weathering have undergone a gradual long-term acidification process.

This process has been recognized at least since the studies of Lundqvist (1925) and has been observed in many geographic areas. In Europe, these areas include Sweden (Digerfeldt, 1972, 1975, 1977; Florin, 1977; Renberg, 1976, 1978; Renberg & Hellberg, 1982; Tolonen, 1972), Finland (Alhonen, 1967; Tynni, 1972; Tolonen, 1967, 1980; Salomaa & Alhonen, 1983; Tolonen et al., 1986), Denmark (Nygaard, 1956; Foged, 1969), northwestern England (Evans, 1970; Haworth, 1969; Round, 1957, 1961; Pennington, 1984), Scotland (Alhonen, 1968; Pennington et al., 1972), Wales (Crabtree, 1969; Evans & Walker, 1979; Walker, 1978), Czechoslovakia (Rehakova, 1983), and Greenland (Foged, 1972). Fewer regions in North America have been investigated, but diatom, chrysophyte, and sediment chemistry data indicate that long-term acidification has occurred in Mirror Lake, New Hampshire (Sherman, 1976, 1985); Cone Pond, New Hampshire (Ford, 1984, 1989); Compton, Quincy, and Bottomless Pit Bogs, and Bear and George Ponds in New Hampshire (Ryan & Kahler, 1987); Bethany Bog, Connecticut (Patrick, 1954); Berry Pond, Massachusetts (Rochester, 1978); Duck Pond, Cape Cod, Massachusetts (Winkler, 1988); Holmes Lake (Rhodes, 1985) Heart Lake, Upper Wallface Pond, and Lake Arnold in the Adirondack Mountains, New York (Reed, 1982; Whitehead et al., 1986; Christie & Smol, 1986); Crystal Lake, Wisconsin (Conger, 1939); Lake Mary, Wisconsin (J.C. Kingston, pers. comm.); Vestaberg Bog, Michigan (Colingsworth et al., 1967); Red Rock Lake, Colorado (Norton & Herrmann, 1980); Green Lakes 5, Niwot Ridge, Colorado (Kerstin Williams, pers. comm); Splan Lake, New Brunswick (Rawlence, 1988); and on Ellesmere Island, above the arctic circle (Smol, 1983). Some lakes have apparently been naturally acidic from the time they first formed (e.g., Round Loch of Glenhead, Jones et al., 1986).

We can make some generalizations based on these studies. The magnitude and the rate of long-term acidification vary among lakes and within lakes. Diatom inferred pH (DI), when it has been calculated, indicates declines from 0.5 pH unit or less to as much as 2.5 pH units; for example, from pH 7.5 to pH 5.0 for Upper Wallface Pond in the Adirondacks (Whitehead et al., 1986) and from pH 7.0 to 6.0 for L. Gårdsjön, southwestern Sweden (Renberg & Hellberg, 1982). Rates of change are gradual; declines of 1 pH unit take hundreds to thousands of years. In general, lakes with the highest current pH have acidified the least, and data for lakes with pH

currently above about 7.5 indicate little or no long-term acidification [e.g., Linsley Pond (Patrick, 1943), Pickerel Lake (Haworth, 1972), Sunfish Lake (Sreenivasa & Duthie, 1973), Kirchner Marsh (Brugam, 1980), South King Pond (Ford, 1984, 1989) Little Round Lake (Smol & Boucherle, 1985)].

The most rapid acidification generally occurred during the early postglacial period. Following deglaciation, the lakes were commonly alkaline (pH 7.5 or above), with the major reason for the high pH being the presence of unweathered till and outwash deposits with large supplies of easily leachable base cations. Alkalinity declined after easily leached bases were removed and as soil and vegetation developed. These processes have been shown to be important in soil and lake development (Crocker & Major, 1955; Livingstone et al., 1958; Andersen, 1966; Berglund & Malmer, 1971; Dillon & Evans, 1982) and seem to recur during each interglacial period (Iverson, 1958; Andersen, 1966, 1967; Miller 1986). The extent of the decline in pH and alkalinity depended largely on the characteristics of the soil, bedrock, climate, and vegetation. Acidification was greater in regions where geologic materials are more resistant to weathering, where soils and glacial deposits are thinner, where rainfall is greater, and where vegetation results in greater output of organic acids. The early period of most rapid acidification was typically followed by long periods of relative pH stability or very gradual decline (usually 1.0 pH unit or less). Some fluctuations in pH occurred and are attributed to changes in factors such as climate, watershed vegetation, and lake level changes. For European lakes especially, other events such as seawater intrusion (connection with saltwater environments) and land use changes caused by early inhabitants also affected lake water pH.

2. Watershed Disturbance

Logging, fire, blowdown, and similar sudden disturbances can change lake pH and alkalinity, although there is evidence that these changes are often relatively minor and short lived (Gorham et al., 1979; Wright, 1981; Nilsson et al., 1982; Martin et al., 1984). Several factors can affect the response of a lake to watershed disturbance and determine whether pH increases or decreases and

by how much and how fast. These include (1) the nature, extent, and severity of the disturbance, (2) the past frequency of similar disturbances, (3) the influence of drainage pattern and density, geology, soil type and texture, steepness of watershed slopes, and hydrology, and (4) taxonomic composition and rate of vegetation regrowth.

One well documented response to disturbance is the initial decrease in pH following logging of watersheds at Hubbard Brook. This decrease was caused by increased streamwater nitrate concentrations resulting from mineralization of soil organic matter and reduced slant uptake (Bermann & Likens, 1977). Another possible pattern of pH change would begin with an initial, rapid increase in pH as the flux of cations released from a watershed increased. This would be followed by a gradual decline to below predisturbance pH as the cation flux decreased because of landscape stabilization and increased uptake by rapidly regrowing vegetation (Nilsson et al., 1982; Gorham et al., 1979). The loss of a forest canopy could change the type and amount of dry deposition, thereby altering the depositional flux of acidifying compounds. Alternatively, reforestation could lead to increased scavenging of aerosols and therefore increased S loading (Harriman & Wells, 1985). More gradual land use change, such as the decline in certain agricultural practices in Scandinavia in the past 100-200 years might also cause lake acidification. According to Rosenqvist (1978), the regeneration of natural vegetation would lead to an increase in soil humus content and ion exchange capacity, removing more cations from soil water, which would result in surface water acidification. Over an even longer period of hundreds to thousands of years, the change of forest to heathland, particularly in western Europe, might have affected the proton flux to lakes and pools.

Detecting the effects of watershed disturbance in the sediment record can be difficult if water chemistry changes are short-lived. This is likely if the sediment interval(s) representing the time of disturbance time was not analyzed or if altered diatom and chrysophyte assemblage composition was not discernible because of sediment mixing or some other factor.

3. Atmospheric Deposition of Strong Acids

Precipitation containing excess strong acids is falling on large areas of eastern North America (Barrie, 1984) and western Europe (EMEP/CCC, 1984). Available evidence suggests that the quantities of strong acids now being deposited are significantly greater than they were before the year 1850 and that this precipitation can cause acidification of lakes (e.g., NRC, 1986; Altshuller & Linthurst, 1984; Odén, 1968; Brimblecone & Stedman, 1982; Ministry of Agriculture, 1982; Schwela, 1983; Asman et al., 1988). Some authors suggest, for example, that the concentration of sulfate in many Adirondack lakes may be five times higher than before 1800 (Galloway et al., 1983; Holdren et al., 1984).

4. Evaluating Causes of Recent Acidification

Causes of lake acidification can be evaluated using paleoecological data to test alternative hypotheses. Studies can involve single lakes, or comparisons of sites that have, for example, high and low acid deposition, or that are forested or not. First, the logical consequences of each hypothesis are specified in terms of trends that can be observed in sediment cores. Then, these are compared with measured trends in sediment cores in attempts to demonstrate that the logical consequences do not occur, and therefore that the hypothesis is falsified (Charles & Whitehead, 1986a; Battarbee et al., 1985a; Jones et al., 1986).

a. Natural Long-Term Acidification

Recent declines in lake pH caused by natural, long-term acidification processes should be continual throughout the period studied. Based on studies of long-term lake histories, the overall decline should be small, no more than about 0.1 pH unit in a 20-year period. There should be no major changes in taxa composition or at least no major shifts in the percentage of diatom

valves assigned to each pH category. There should be no correlation between DI pH changes and indicators of watershed disturbance or acidic deposition.

b. Watershed Disturbance

If a pH decline was caused only by watershed disturbance and processes associated with vegetation recovery, the period of pH changes should logically be related to the time of the disturbance; the larger and more rapid the pH changes, the closer they should have been to the time of disturbance. A pattern that might be expected is a sudden change in pH followed by a gradual return to previous conditions, although there are few data to substantiate this contention. In the case of fire by itself, or in combination with logging, a few studies suggest there may be an increase in pH after the disturbance, followed by a gradual decrease (Dickman & Fortescue, 1984; Tolonen et al., 1986; Davis et al., 1989). In Scotland, Flower & Battarbee (1983) and Flower et al. (1987) were able to falsify the watershed change hypothesis by showing that acidification of some lakes began well before the watersheds were planted with coniferous trees. In the Adirondacks, New York, lake acidification began before a major blowdown event (Johnson et al., 1987).

In addition to changing the pH, watershed disturbance may affect the nutrient input to lakes. This effect alone can cause changes in the composition of diatom assemblages. In The Netherlands, changes in human impact (e.g., discontinuation of sheep washing in moorland pools) plays a role in acidification (Van Dam et al., 1988; Van Dam, in press a).

c. Atmospheric Deposition of Strong Acids

Lake acidification can be attributed to acid deposition if (1) the lake is in a region receiving deposition of excess strong acids and there is evidence of industrial pollution in the upper levels of the sediment, (2) DI pH declines occur after an increase in acid deposition could reasonably be expected to have taken place, based on knowledge of current water chemistry and watershed

acidification processes, and (4) changes are not correlated with, and cannot be attributed to, watershed changes or other local factors.

When these criteria are used to evaluate causes of acidification, several factors must be emphasized. First, more than one of the above processes may be affecting a lake simultaneously. Second, the history of each lake watershed system and nearby emission sources should be investigated thoroughly, so that all possible causes of acidification can be evaluated. Third, interpretations based on diatom and chrysophyte data should be consistent with interpretations of other sediment data, and current models of lake acidification processes. Fourth, assessments of regional acidification trends should be based on studies of several lakes within a region.

III. Assessment of Recent Lake Acidification Trends in Eastern North America and Europe

A. Selection and Analysis of Data Sets

Diatom and chrysophyte assemblage data were chosen using the following criteria as guidelines: (1) present lakewater alkalinity less than 200 μeq/L (acid neutralizing capacity low enough so that lake pH and diatom assemblage composition could have been altered as a result of significant change in watershed characteristics or atmospheric deposition of acids; some lakes with higher alkalinity chosen as reference lakes that should have had stable pH in recent years), (2) minimal or, if not minimal, well-known watershed disturbance or influence from local emission sources, industrial effluents, or other factors, (3) sufficient quality and quantity of diatom or chrysophyte data, with taxonomic identification to lowest possible level, an adequate number of time intervals analyzed, and access to primary data including counts, (4) sufficient ecological information on at least the most common diatom taxa, including distribution with lakewater pH within the region, (5) quality of dating, and (6) the availability of data for other lake, watershed, and sediment characteristics related to acid deposition and lake acidification.

The steps in analyzing the data were as follows: (1) assemble all relevant data sets, (2) evaluate pH and acidification trends by type of lake and by geographic region, (3) evaluate

potential causes of the trends, and (4) draw conclusions concerning trends in acidic deposition. Most of the lakes discussed in this chapter are listed in Tables 1 and 2. Many of the data are as yet unpublished because investigators have produced the data only recently.

Summaries of research results for the major regions are presented in varying formats. This largely reflects differences in the amount, quality, and consistency of data for making regional assessments, the extent to which the data have already been synthesized, and the most relevant issues that need to be addressed.

Inferred pH values are based on calibration sets developed for regions in which the study lakes are located. The calibration data and ecological analyses of diatom and chrysophyte taxa are summarized or referred to in the various papers cited in the sections.

B. North America

1. United States

To evaluate recent acidification trends in North America, diatom data sets from several regions (Figures 2 and 3; Table 1), meeting the minimal criteria listed in the section above, were assembled and reviewed. Most of the lakes in the Adirondacks (12), Upper Midwest (9), and northern Florida (6), and some in New England (3), were studied as part of the Paleoecological Investigation of Recent Lake Acidification project (PIRLA) funded by the Electric Power Research Institute (Charles & Whitehead, 1986a).

In general, the lakes are not broadly representative of each region, even of the poorly buffered lakes. Most of the study lakes represent the most acidic lakes in each region. For example, the 14 New England lakes are among the most acidic in the region and 9 of them were among the 20 lowest pH lakes of the 94 lakes studied by Norton et al. (1981), all of which had pH less than 7.0.

Chrysophyte data were available for cores from Big Moose Lake (Smol, 1986), Deep Lake (Smol et al., 1984b), Upper Wallface Pond, Queer Lake, Windfall Pond, Lake Arnold, Little Echo

Pond, and West Pond (J.P. Smol, unpublished) in the Adirondack Mountains; Little Long Pond, Mud Pond, and Haystack Pond in New England (Dixit et al., 1989); and some lakes in the Sierra Nevadas. Major changes in diatom stratigraphy were usually accompanied by changes in chrysophyte stratigraphy and they both indicated the same pH trend.

a. Adirondack Mountains, New York

Diatom and chrysophyte assemblages in Adirondack lakes indicate significant recent acidification (Table 1). All 11 clearwater lakes with current pH < 5.5 have acidified recently. The acidification histories of most are substantiated by several lines of evidence, including stratigraphies of diatom, chrysophyte, chironomid, and Cladoceran remains, Ca:Ti and Mn:Ti ratios, sequentially extracted forms of Al, and historical fish data (Charles et al., 1987, 1989; Charles & Norton, 1986; Davis et al., 1988). The pH of Big Moose Lake, Queer Lake, and Honnedaga Lake (Figure 4) declined about 1 unit from before 1800 to the present. The decrease in the pH of Big Moose Lake and Queer Lake is associated with recent increases in acidobiontic taxa, including Fragilaria acidobiontica Charles and Navicula tenuicephala Hustedt (= ?Stauroneis gracillima Hustedt), and decreases in the indifferent, euplanktonic taxon Cyclotella stelligera Cleve and Grunow. The increase in chrysophyte taxa that indicate acidic conditions, such as Mallomonas hamata Asmund and M. hindonii Nicholls (Smol et al., 1984a,b) also indicates a lowering of pH. Diatoms and chrysophytes in Deep Lake, Barnes Lake, Lake Arnold, Merriam Lake, Upper Wallface Pond, and Woods Lake indicate a recent drop in pH from about 5.0-5.5 to around 4.7-4.9. These lakes may be considered to have been naturally acidic and to have acidified further recently. Shifts in taxonomic composition (Charles, 1986; Charles et al., 1989) suggest that increased concentrations of Al or other metals, and decreased organic matter (Davis et al., 1985, 1988) may have accompanied the pH decline. Acidification trends appear to be continuing in some lakes (e.g., Queer Lake; Figure 4), despite recent reductions in atmospheric sulfur loading.

All clearwater lakes with current pH greater than 6.0 have acidified only slightly or not at all, reflecting the ability of the lake/watershed systems to neutralize acid inputs. Diatoms and

chrysophytes in high DOC lakes and bogs (Little Echo Pond, West Pond), suggest a relatively small pH change, as would be expected in highly colored acidic waters buffered by organic acids. All DI pH values (Table 1), except for Seventh Lake, were calculated using inference equations developed by Charles (1985) and Charles & Smol (1988).

The primary cause of recent acidification trends clearly is the increased atmospheric deposition of strong acids derived from combustion of fossil fuels, though other factors may play a role. Natural processes and watershed disturbances cannot account for the changes in water chemistry that have occurred. For many of the lakes, sediment core profiles of Pb, Ca, V, Zn, S, polycyclic aromatic hydrocarbons, and carbonaceous sphaerules provide a record of deposition of materials associated with fossil fuel combustion beginning in the late 1800s and early 1900s. The onset of acidification began after that time.

b. New England

Changes in sediment diatom assemblage composition in New England lakes indicate recent acidification (Norton et al., 1985); however, most of the pH trend data indicate no change or a slight decrease (e.g., in Speck Pond; Figure 5) in pH from pre-1800 to the present. At Ledge Pond, a total decrease of 0.6 to 0.7 unit, starting slowly during the 1800's but accelerating after 1960, has been inferred (Davis et al., 1983; Norton et al., 1985). Fluctuations in some of the profiles appear to be related to watershed events (Davis et al., 1983). Sediment chemistry data for Mud Pond, Haystack Pond, and Little Long Pond indicate post-1900 increases in materials associated with the combustion of fossil fuels (e.g., total Pb and V) (Davis et al., 1989).

The DI pH of Duck Pond, a kettle pond on Cape Cod, Massachusetts, suggests a slight recent acidification trend (pH 5.2 to 5.0), though this pond has been naturally acidic for thousands of years, with a mean DI pH for its entire history (about 12,000 years) of 5.2 $\pm$ 0.3 pH units.

c. Northern Great Lakes States

Lakes in the Upper Midwest differ as a group from those in the Northeast in that most are seepage lakes and are very dilute (conductivities as low as 9 μS/cm) and low alkalinity (-38 to 80 μeq/L). Diatom inferred pH trends indicate little or no recent acidification. There is no indication of acidification of Minnesota (3 lakes studied) or Michigan lakes (2 lakes studied), but there is evidence of post-1900 pH declines in some Wisconsin lakes (4 lakes studied). McNearney Lake in Michigan was the only naturally acidic lake studied (pre-1800 DI pH = 4.9); its pH has not declined, but Al concentration is high. Concentrations of S, Pb, Cu, V, and polycyclic aromatic hydrocarbons (PAH) increase towards the surface of the cores, but they are not as great as concentrations in the northeastern United States (Kingston et al., 1989).

d. Florida

Northern Florida has the largest percentage of lakes with pH < 5.0 (12%) of any region in the United States (Landers et al., 1988). Analysis of pre-1850 sediment core diatom assemblages indicates that four of six lakes had low pH values due to natural causes (Table 1; Sweets et al., 1989). Low cation concentrations are probably attributable to the low cation exchange capacity and base saturation of the deep coarse sands in which the lakes are situated, and isolation from the local groundwater table (Floridan aquifer) by the clay-rich Hawthorne formation. The high SO_4^{2-} concentrations, which contribute most to the acidity, apparently result from high evapoconcentration rates (Pollman & Canfield, 1989).

Lake Barco and Lake Suggs have become more acidic recently, with DI pH decreases of 0.7-0.9 pH units. There are two possible causes: (1) acidic deposition based on the timing of pH changes and the analysis of sediment chemistry, and (2) a decrease in the regional groundwater table, as shown by dynamic lake models (Sweets et al., 1989; Pollman & Canfield, 1989).

e. Rocky Mountains and Sierra Nevada

The Rocky Mountain and Sierra Nevada lakes may be considered control or reference lakes. They all have low alkalinity (most with alkalinity < 60 μeq/L) and are therefore sensitive to increased input of strong acids, but do not currently receive precipitation with an annual average pH lower than about 5.0 (Gibson et al., 1984; Melack et al., 1987). Current SO_4^{2-} concentrations are typically < 5 μeq/L. These lakes are considerably more dilute, and potentially more likely to acidify, than lakes in the Eastern United States (Eilers et al., 1988). Changes in the diatom stratigraphy of most Rocky Mountain lakes are relatively minor, occur gradually, and indicate no sign of acidification (Baron et al., 1986). The same is true for Sierra Nevada lakes (e.g., Holmes, 1986), but there is evidence of trends of both increasing (e.g., Emerald Lake) and decreasing pH (Lake 45), particularly based on analysis of floristic changes (M. Whiting, pers. comm.). Some of the trends began well before 1900. Causes are not clear, but may include effects of volcanic eruptions during the past few thousand years, changes in dry deposition of particulates containing cations because of land use changes in nearby valleys, and increased episodic acidification events following summertime rainstorms (M. Whiting & D. Whitehead, pers. comm.).

f. Ongoing Studies

Studies of surface sediments and cores are being conducted on lakes of the Pocono Plateau in Pennsylvania (John Sherman, Academy of Natural Sciences of Philadelphia, pers. comm.). Marjorie Winkler (U. of Wisconsin) is analyzing several short (0.5-1.0 m) and long cores from lakes on Cape Cod, Massachusetts. Mark Whiting, Donald Whitehead, Aruna Dixit, and John Smol are studying several more lakes in the Sierra Nevadas. John Smol, Donald Charles, John Kingston, Sushil Dixit, Keith Camburn, and Brian Cumming are analyzing tops and bottoms of cores from about 40 randomly selected Adirondack lakes (studied as part of the EPA Eastern Lake Survey, Phase I, Landers et al., 1988). Kerstin Williams, (Institute of Arctic and Alpine Research), is studying

long-term trends of Blue Lake and Green Lakes 4 and 5 at the Niwot Ridge Long-Term Ecological Research (LTER) site in Colorado.

2. Eastern Canada

A number of investigators, usually working independently, have used paleoecological techniques to study acidification trends in a wide variety of lakes in Eastern Canada. In contrast to the United States, where the goal of most studies has been to assess the effects of acidic substances transported long distances, a large percentage of the Canadian studies focus on lakes that are near large point sources of SO_x and metal emissions (e.g., Sudbury, Wawa), or near mining effluents, or on lakes that have been manipulated (e.g., limed or acidified). In addition, several studies deal with lakes that are strongly influenced by organically rich soils and bogs. Nonetheless, in all lake regions studied thus far where acidic precipitation is falling, diatoms and chrysophytes have indicated recent pH decreases in at least some lakes. The taxonomic changes are similar to those recorded in the northern United States and some areas of Europe.

a. Ontario

Using Index α, calibrated with data for 28 lakes located north of Lake Superior in the Algoma District near Wawa, Ontario (Dickman et al., 1984; Dixit, 1986a; Dixit & Dickman, 1986) Dixit (1983) inferred that the pH in Lake CS had markedly declined, that Lake X4 has been rather acidic for the last 200 years, that Lake U3 has had a relatively stable pH over recent history, and that Lake W1 has had a fluctuating pH during its recent history. The acidification of Lake CS and another lake, B, in the past 30-50 years is apparently attributable to local emission of S near Wawa, Ontario, in the 1940's and 1950's (Dickman et al., 1984; Dickman & Fortescue, 1984; Somers & Harvey, 1984). Forest fires and logging may have affected diatom stratigraphy and lakewater pH. Dickman & Thode (1985) provided profiles of DI pH for four additional lakes in the Wawa area: Logger, Beaver, WW1, WW2. Beaver Lake has acidified considerably; WW1 and

WWII are located in a carbonate-rich greenstone area and did not acidify, nor did Logger Lake, which is located 100 km upwind of the emission sources and served as a control.]

In Batchawana Lake (Delorme et al., 1986), a slightly acidic lake located northeast of Lake Superior, diatom changes predate cultural activities in the area, and are interpreted as being caused by climatic changes. There is no evidence of any acidification trend over the past 150 years.

Taylor (1986) studied the diatom assemblages in the surficial sediments of 60 poorly buffered lakes in central Ontario. Using multivariate statistics, lakewater pH and humic content were identified as the two most important chemical variables affecting diatom distributions in his calibration set. The humic content of lakes has influenced the accuracy of these equations; also physical factors and some metal factors were stressed.

Dixit (1986b; Dixit & Evans, 1986; Dixit et al., 1987a, 1988a,b) used diatoms and chrysophytes and a 30-lake calibration set to infer the pH history of Hannah and Clearwater Lakes. In Hannah Lake, acidification started soon after the roasting of ore began at Copper Cliff in the 1880's. Between about 1880 and 1975, the DI pH declined from about 6.0 to about 4.6. However, after liming in 1975, the DI pH increased. In Clearwater Lake, the diatom assemblages indicated that acidification occurred after the installation of tall stacks at Copper Cliff in the 1920s. The pH declined from about 6.0 in ca. 1930 to about 4.2 by ca. 1970. Acidification appears to have stopped after that time.

Chrysophyte scale remains from a variety of Ontario lakes are being used to infer pH change (K. Nicholls, Ontario Ministry of the Environment, and J.P. Smol, unpublished data). Chrysophytes indicate a decrease in lakewater pH in Algonquin Park (Maggie and Pincher Lakes), in the Parry Sound region (Lady and Raven Lakes), and in Pukaskwa National Park (two unnamed lakes). The pH declines are currently being quantified.

Quirke Lake (near Elliot Lake, Ontario) apparently acidified as a result of local mining and milling of uranium, although acid precipitation may also have been involved (McKee et al., 1987). The pH declined in the 1950's and reached a minimum in the early 1960's, when it dropped to about 4.5 to 5.0. With the collapse in the uranium market, the mine closed and the diatoms recorded the recovery of the lake to its present pH of 6.4.

Davidson (1984) studied artificially acidified Lake 223 in the Experimental Lake Area using a variety of techniques (e.g., tape peels and development of pH inference techniques based on a calibration set of plankton samples taken from the lake during the period of artificial acidification). The diatom flora in a core from the lake showed good agreement with the known plankton history. These results further document the rapid response of diatom flora to changing water chemistry, and indicate that the changes are recorded in the sediment. Dickman et al. (1988) also studied a core from this lake and found that diatom flora shifts and inferred pH trends correlated with the acidification trend.

b. Quebec

Cores from Key Lake (pH = 5.2) and Lake C-22 (pH = 5.8), in the Matamek Watershed in northeastern Quebec, were studied by Hudon et al. (1986). In Key Lake's recent sediments there was a decrease in the number of indifferent taxa and an increase in acidobiontic diatoms; however, no statistical increase in acidity could be inferred. Similarly, in Lake C-22 there is a slight tendency to greater acidification in the last 20 years, but the trend is less distinct than that shown in Key Lake.

Dixit et al. (1987b, 1988c, 1988d) have conducted paleolimnological work (diatoms, chrysophytes, geochemistry, Pb-210 dating) on a series of LRTAP (Long Range Transport of Air Pollutants) lakes in Quebec. Located in a strip 150 km wide north of the St. Lawrence River between the Ottawa and Saguenay rivers, the 25 lakes chosen for this study have been extensively monitored by the Inland Waters Directorate. All the lakes are remote. In most of the study lakes, there was either a small change or no change at all in inferred pH. Four of the lakes suggested a recent minor pH reduction. Interestingly, chrysophytes appeared to change much more strikingly in recent history in several of the study lakes (e.g., Figure 6); but again suggesting only minor acidification.

c. Atlantic Provinces

Vaughan et al. (1982) compared surface sediment diatom assemblages of four Halifax, Nova Scotia, area lakes with assemblages recorded by G.H. Yezdani (unpublished) in the same lakes in 1971. No attempt was made to quantitatively reconstruct past lakewater chemistry, but Vaughan et al. (1982) showed that there was a marked relative increase in acidophilic and acidobiontic diatom taxa in the 1980 samples, as compared to those recorded by Yezdani in 1971.

Delorme et al. (1983) inferred recent DI pH trends for Kejimkujik Lake (central Nova Scotia), a presently acidic (pH = 4.8) site in a drainage basin of organic rich soils and bogs, overlying weathering resistant igneous and metamorphic rocks. Since those results were published, the core has been reanalyzed using more recent taxonomic information. Instead of a significant increase in DI pH, smaller trends are now inferred (H. Duthie, pers. comm.). The lake receives wet precipitation with an average weighted mean pH of 4.6. Cultural impact in the drainage began in the mid-1800's with activities such as deforestation and the burning of lumber refuse. The present low pH of the lake cannot be attributed entirely to atmospheric loading; acidic conditions have persisted for at least the last 1000 years, probably because of organic acids.

Walker & Paterson (1986) described the distribution of diatoms in the surficial sediments of 22 lakes and 6 peatland pools from eastern New Brunswick and adjacent Nova Scotia. This study provides data pertinent to natural acidification processes in Atlantic Canada and to paleoecological studies of recent, anthropogenic acidification. Calibrations based on Index B and multiple regressions were performed. The peatland pool data were not used in the calibration because they did not appear to conform to the general trend of the pH log index B relationship based on the other lakes.

A study of three Nova Scotia lakes and four New Brunswick lakes showed DI pH declines in the unbuffered lakes (e.g., a decline from 6.1 to 5.3 for Big Indian Lake), but not in the buffered lakes (Elner & Ray, 1987). The changes occurred over the last 70 or so years, coincident with assumed increases in acidic precipitation.

Scruton & Elner (1986) and Scruton et al. (1987a,b) have completed a diatom calibration for 34 lakes and DI pH profiles for 7 lakes in Newfoundland. The Index B inferred pH suggested declines of about 0.2-0.3 pH unit in three of the lakes. The pH changes are presumed to be caused by atmospheric sources, as well as by possible watershed disturbances (e.g., forest fire). Humification is also possible. The slight inferred pH changes are consistent with the relatively low level of acidic deposition in Newfoundland.

C. Europe

1. United Kingdom

In the United Kingdom, there has been an intensive study of the extent and causes of lake acidification using paleolimnological techniques (Flower & Battarbee, 1983; Battarbee, 1984; Battarbee et al., 1985a,b; Jones et al., 1986; Flower et al., 1987, 1988; Battarbee, 1988a). The results as of early 1988 for the United Kingdom as a whole are described in Battarbee et al. (1988, Report). Figure 7 shows the sites studied and Table 1 summarizes the data.

a. Diatom Trends

Diatom analysis of the recent sediments of acidified lakes in the United Kingdom shows that in almost all cases the preacidification (before 1850) floras of these lakes were remarkably stable. The same species occurred in approximately the same proportions at each site for hundreds of years, despite shifting land-management patterns and climatic changes.

At many sites during the preacidification period, a significant proportion of the diatom assemblage was composed of planktonic forms, especially Cyclotella kützingiana Thwaites. However, at some sites, plankton was absent or rare despite apparently stable conditions and a relatively high pH. So far, the reasons for this difference between sites are not known, although auxiliary factors such as water depth and nutrient availability may be important.

In cores from all acidified sites studied, C. kützingiana, where originally present, is the first species to decline. In most cases (e.g., Llyn Hir, Loch Laidon, Scoat Tarn), the decline is abrupt and absolute.

The nonplanktonic flora at most sites prior to acidification are dominated by Achnanthes minutissima Kütz, Anomoeoneis vitrea (Grun.) Ross and Fragilaria virescens Ralfs (e.g., Figure 8). These species have mainly circumneutral pH distributions, but are also found in abundance mixed with more acidophilous forms between pH 5.5 and 7.0.

As the sensitive circumneutral species decline, a proportionate increase in acidophilous species occurs. These species are invariably already present in reasonable abundance in the pre-acidification flora and include Tabellaria flocculosa (Roth) Kütz., Frustulia rhomboides (Ehr.) Det., and varieties, Eunotia veneris (Kg.) O. Müll., (= E. incisa (W. Smith) ex. Greg.), Achnanthes marginulata Grun., and Navicula heimansii van Dam & Kooyman (= N. leptostriata Jørgensen).

Further acidification leads to the elimination of circumneutral taxa, to the continued expansion of some acidophilous species, and to the appearance and increase of acidobiontic taxa such as Tabellaria quadriseptata Knud., and T. binalis (Ehr.) Grun. However, the response of any one site is variable.

At the two higher pH sites chosen for analysis, Loch Urr (pH 6.8) in Galloway and Burnmoor Tarn (pH 6.4) in Cumbria (Table 1), only slight changes in diatom assemblages have been recorded and, in both cases, the flora are dominated by planktonic taxa, including C. kuetzingiana and circumneutral nonplanktonic taxa, especially Achnanthes minutissima, and Anomoeoneis vitrea.

b. pH Trends and Extent of Surface Water Acidification

At all sites except Loch Urr and Burnmoor Tarn, a clear post-1850 acidification is indicated by the trends in diatom assemblages. Quantification of these trends using the multiple regression approach (Flower, 1987) gives results in good agreement with measured pH for modern day samples (Table 1). However, at some sites (e.g., Llyn Hir) a full reconstruction is not possible

because of the numerical importance of unknown species. In this case, the approximate pH change can be inferred from the difference between the diatom-based pH for the preacidification period and the measured pH of today.

At most of the acidified sites, those with present pH levels < 5.0 had pH levels about 6.0 prior to 1850. Only two sites, Loch Enoch and Loch Tanna, had pH values < 5.5 at that time. These data show that sites with zero or negative alkalinity prior to 1850 or so were exceptionally rare.

The largest inferred pH declines occurred in the Welsh lakes in the Rhinog area. Here pH has probably decreased by 1.0 to 1.5 pH units. These lakes have extremely clear water (TOC < 1 mg/L) and the lack of a significant organic acid input from the catchment was probably the reason for their relatively high preacidification pH. In general, pH declines since 1850 have varied from about 0.5 to 1.5 pH units, depending on the initial sensitivity of the lake to acidification and the intensity of acid deposition.

Although sensitive areas in Scotland, Northern England, and Wales have been acidified to a similar extent and over the same broad time period (since 1850), there is considerable variation among sites in the timing of the change and in the acidification trajectory (Figure 9).

In Wales, acidification began in the mid- to late-19th century at Llyn Hir, Llyn Dulyn, and Llyn Llagi, in the early 20th century at Llyn y Bi, but not until the mid-1940's at Llyn Eiddew Bach and Llyn Gynon. In Scotland, acidification began at many sites in the mid-19th century, but at other sites it was delayed until the turn of the century (e.g., Loch Dee) or later (Loch Grannoch and Loch Chon). In Cumbria, some upland tarns started to acidify in the mid-19th century (Scoat Tarn), but other sites (e.g., Greendale Tarn) did not begin to change until about 1900. And at some sites, although the first sign of acidification occurred in the mid-19th century, an acceleration in the rate of acidification occurred after 1940 (e.g., Llyn Llagi).

Although we have as yet insufficient catchment information to explain in detail why this kind of variation has occurred, it is clear that the response of any site depends on the historical pattern and intensity of acid deposition and the general neutralizing capacity of the catchment soils and groundwater.

The most sensitive sites (Loch Enoch, Loch Valley, Lochnagar, all with Ca^{++} < 35 μeq/L) appear to have acidified continuously from about 1850 until around the present time (Figure 9). At less sensitive sites (Loch Dee, Loch Tinker, with Ca^{++} 50-80 μeq/L) or at sites with lower acid deposition (Loch Laidon, with Ca^{++} 40 μeq/L), the acidification has been slow or has been delayed, and, in the case of Burnmoor Tarn and Loch Urr, with Ca^{++} > 150 μeq/L, acidification has not occurred despite their location in areas of high acidic deposition (Battarbee et al., 1988c).

c. Extent of Surface Water Acidification

Earlier work in Galloway, Southwest Scotland, showed that recent (post-1850) acidification had occurred at lakes lying on the two upland granitic areas in that region (Flower & Battarbee, 1983; Battarbee et al., 1985a; Flower et al., 1987). The extension of this work to other regions of the United Kingdom shows that similar acidification has occurred at lakes in central and north Wales on Lower Palaeozoic sedimentary and metamorphic rocks, in Cumbria on Borrowdale volcanic strata, and throughout Scotland southeast of Loch Ness on granites. Exceptionally acidic waters also occur in the English Pennines. Although sufficiently old sediments are not available for analysis in this region, it can confidently be predicted that these waters are also acidified.

Only a small number of water bodies have been analysed in each of the regions of the United Kingdom considered. However, the patterns are so consistent, both among and within regions, that this interpretation can be extrapolated confidently to other water bodies with similar modern water chemistry in these areas. Although many of the sites are quite remote, almost all are of special interest because they are situated in National Parks, National Nature Reserves, Sites of Special Scientific Interest, Man and Biosphere Reserves, and National Trust areas.

The full geographical extent of acidification is not yet known. Work in progress (Figure 7) is designed to reconstruct the recent history of sensitive sites in the northwest of Scotland, in Ireland, and in parts of lowland England. Preliminary data suggest that little acidification has taken place in northwest Scotland and western Ireland, but that in eastern Ireland there may be acidified lakes in the Mourne and Wicklow mountains.

d. Causes of Surface Water Acidification

It has already been established that the primary cause of lake acidification in Galloway is acid deposition (Battarbee et al., 1985a; Jones et al., 1986, 1988). More recent data from other parts of Scotland, Wales, and England (Battarbee et al., 1988a,b) support this conclusion. Not only is the evidence consistent with this hypothesis, but also the overall pattern of observations cannot be accounted for by alternative hypotheses. There are a number of cases where recent (post-1950) conifer afforestation has played a role, but its effect is variable from site to site.

The evidence for the effects of acid deposition is as follows (Battarbee et al., 1988a):

1. At all acidified sites, the beginning of acidification is consistent with the expansion of industry in Britain from the late 18th to the early 19th century. At no sites did acidification begin before the first evidence of trace metal contamination.

2. Cores from all acidified sites have high concentrations of carbonaceous particles that are derived from fossil fuel combustion.

3. Although no cores from areas of very low acidic deposition have yet been analysed, sensitive sites in northwest Scotland, such as Loch Corrie nan Arr and Lochan Dubh (Ca^{++} < 50 μeq/L), have pH values > 5.5 and the only lochs with pH < 5.0 in this rather low deposition area are shallow, brown-water sites (TOC > 8 mg/L).

4. Although there have been changes in burning and grazing regimes in many of the catchments studied, this has led in most cases to a decline in heathland vegetation rather than to an increase. In Llyn y Bi, the only case with a well-documented increase in catchment Calluna, acidification began before the no-burning policy was implemented when the lake became National Trust property. In any event, it has been

clearly shown elsewhere that vegetation change of this kind cannot be used to explain intense surface water acidification (Jones et al., 1986).

5. There is no evidence that liming has ever taken place in the catchments of any of the sites studied. Cessation of that practice may therefore be excluded as a potential acidification mechanism.

6. Acidification at Llyn cwm Mynach, a site with an afforested catchment, began before afforestation. This was also the case at sensitive Galloway sites (Flower et al., 1987).

The evidence for the additional effects of afforestation is:

1. At Llyn Berwyn, documentary evidence and limited core data suggest that soil disturbance following deep drainage not only caused soil erosion and inwash but also acidification and damage to fish stocks.

2. At Loch Fleet (Anderson et al., 1986), acidification followed afforestation. At this unusual site, acidification may be due to a combination of factors, including the scavenging effect of the forest and the erosion of peat following land drainage. The latter process caused a thick layer of acid organic peat-derived sediment to be deposited across the bed of the loch and this may have sealed the lake from a source of calcium-rich groundwater.

3. Afforestation can cause acidification indirectly through a scavenging mechanism. At Loch Chon, afforestation occurred in the 1950's. Because planting was carried out on the steep catchment slopes by hand, soil disturbance did not occur. However, acidification accelerated about 15 years after planting, although no further

acidification occurred at this time at Loch Tinker, a control site with a moorland catchment.

e. Reversibility

Where sediment accumulation rates are sufficiently rapid, contemporary trends in acidity can be monitored by repeat coring of the lakes described here. Already the uppermost sediments of some Scottish lakes show slight signs of improvement (Battarbee et al., 1988a; Neal et al., 1988), probably in response to post-1970 reductions in acidic deposition in Scotland (Harriman & Wells, 1985). These preliminary data indicate that a large reduction in acidic deposition is likely to lead to a rapid response in lake chemistry and biota.

2. Fennoscandia

Diatom analysis of sediment cores for the assessment of recent lake acidification history was initiated comparatively early in Sweden and Norway, in the early 1970's , as a response to the then growing debate and concern that acidic precipitation may have been causing extensive damage to aquatic environments. Miller (1973) in Sweden, and Berge (1975) in Norway, were pioneers of the technique, and both applied methods developed by Hustedt (1939), Nygaard (1956), and Merilåinen (1967). These investigations were followed by several others. Figure 10 shows the locations of the lakes studied; pre-1800 and current diatom inferred pH values are in Table 1.

a. Norway

Berge (1975) found no recent acidification of Langtjern in central Norway, but in a later study (Berge, 1979) he observed a clear recent acidification in Övre Målmesvatn, a lake in southwest Norway, together with indications of acidification in two lakes where only core tops and bottoms

were analysed. He has also presented ecological data on important diatom taxa that support his inferrences from analyses of sediment cores (Berge 1975, 1985).

R.B. Davis and coworkers have, over a number of years, built up a calibration set for Norwegian lakes. For the purpose of paleoecological inference, they developed three regressions of the diatom data on pH: (1) multiple regression of the Hustedt pH groups, (2) stepwise multiple regression of 13 clusters of the 135 most abundant taxa, and (3) multiple regression of the loadings on the first principal component of 22 abundant taxa (see Davis, 1987). Two regressions of diatom data on TOC were also developed (Davis et al., 1985). Reconstruction of pH on cores from nine lakes has been carried out (Davis & Berge, 1980, Davis et al., 1980, Davis et al., 1983). Five of these lakes show recent acidification; the magnitude of pH decrease is up to 1 pH unit and the timing of the first change is between about 1850 and 1930 (e.g., Figure 11).

Within Norway, investigations of six lakes are in progress as part of the Surface Water Acidification Programme (SWAP) (Figure 11). All are recently acidified except one lake in middle Norway, which is situated in what is considered to be a clean air region.

The number of Norwegian lakes studied is too small to draw any firm conclusions about the geographic pattern of acidification; however, most recently acidified lakes are located in areas with high percentages of acidic lakes that receive high loadings of acidic precipitation.

b. Sweden

Paleolimnological investigations have been made on only five lakes. All are situated in an area of southwest Sweden that is severely affected by surface water acidification. All of the lakes show clear evidence for recent acidification of about 1.5 pH units, based on the sedimentary diatom record.

Miller (1973) [see also Almer et al. (1974)] analysed a core from Stora Skarsjön in southwest Sweden. She found that the abundance of planktonic species had decreased towards the sediment surface and that acidobiontic species such as Tabellaria binalis, Semiorbis hemicyclus (Ehr.) Patr.,

and the Eunotia species had increased. Using index , she estimated that the pH had decreased from about 6 to 4.5.

Renberg & Hellberg (1982) studied three lakes from the same area: Gårdsjön, Härsvatten, and Lysevatten. The subfossil diatom assemblages showed dramatic changes towards the surface in these short cores, from the dominance of circumneutral planktonic species such as Cyclotella kuetzingiana to acidophilous and acidobiontic periphytic species (Figure 12). Index B and a transfer function obtained from a calibration set of 30 lakes were used to infer past pH, which had decreased from about 6.0 to about 4.5 during the post-war period. The dating was obtained by comparing the diatom and oil soot records of the sediments (Renberg & Wik, 1985). Lysevatten, which was limed in 1974, has had a different recent pH history, reflecting the changes caused by the lime treatment. Renberg & Wallin (1985) discussed the results from Gårdsjön further, in particular the problem of sediment record response to rapid pH changes, such as those associated with the acidification process, and also possible causes of the acidification of the lake. Acidic precipitation is believed to be the most likely cause.

Another lake, Lilla Öresjön, a little farther to the south, has now been analyzed as part of SWAP, and the recent trends there are similar to the results from the other lakes.

f. Finland

Paleolimnological research concerning the effects of lake acidification in Finland has undergone a rapid expansion recently. In addition to the core studies briefly reviewed here, the relationships between water quality and sedimentary parameters, including those between pH and diatoms, is being investigated by Meriläinen, Huttunen and others (e.g., Huttunen & Meriläinen, 1983, 1986a,b; Christie et al., 1988; Oksanen et al., 1988).

The results of the anlayses of more than 50 sediment cores from Finland show that recent acidification is widespread throughout the country. Dating seems to indicate that the acidification started later in Finland than in Norway.

Tolonen & Jaakkola (1983) found that three lakes near Helsinki in southern Finland had all acidified by about 1 pH unit. Dating by ^{210}Pb indicated that the acidification started recently, during the 1960's (Hauklampi) or later.

Simola et al. (1985) investigated the diatom stratigraphy of cores from 10 lakes dated using ^{210}Pb; pH was inferred using Hustedt's pH categories and index B. On the basis of their pH development the lakes fell into three groups. The first group contained three lakes in which the pH had declined by 0.3-0.8 pH units from reasonably steady background level. According to ^{210}Pb dating, the decline in two of these lakes had occurred since the 1960s. The second group, also composed of three lakes, showed a similar recent decline, but in these lakes the oldest DI pH obtained was similar to that near the surface. These trends were interpreted as a re-acidification after a period of cultural influence that had caused pH to increase. The third group consisted of four lakes. One was limed, two showed no significant inferred pH change, and one had badly corroded diatoms.

Tolonen et al. (1986) analysed the diatom records in sediments from 10 forest lakes. A striking decline in DI pH (Index B, Index α) occurred in 6 of those during about the last 30 years. Analysis of chrysophycean scales in one of the lakes (Munajärvi) supported the results of the diatom analysis. In some cases, atmospheric deposition was considered the most likely cause of acidification, but in most cases, effects of land use changes could not be ruled out as important factors.

Work is in progress on a large number of lakes in Finland (Table 1, Figure 10), and preliminary results have kindly been made available to us. The investigations include a study by Tolonen and Suksi (pers. comm.) of 3 lakes that have all recently acidified, a study of 24 lakes by M. Liukkonen (pers. comm.), and a study of six lakes by J. Turkia (pers. comm.). About 50% of the lakes in the two latter studies show indications of recent acidification.

3. Western and Central Europe

The small-scaled landscape of central Europe has been cultivated for a very long time. Consequently, the region no longer contains any pristine water bodies, although there are several relatively undisturbed small lakes and pools in nature reserves. Because past human activities (e.g., agricultural drainage water, foddering of ducks and fishes) in and near pools and lakes are not always well documented, it is often difficult to determine the relative importance of land use changes versus acidic deposition in causing DI pH trends.

The majority of the acid sensitive water bodies in the North Sea lowlands are moorland pools. The open water bodies in many of these pools were formed by excavation of peat. Most moorland pools are seepage pools with a perched water table. Consequently, the catchment area is only slightly larger than the area of the water surface. Nevertheless, land use around these pools can have a profound influence on their water quality (e.g., the litter of trees falling in the pools or the washing of sheep in the pools). The problems associated with paleoecological investigations of these shallow waters, such as wind mixing, bioturbation, desiccation, and presence of a dense aquatic vegetation, have been discussed by Dickman et al. (1987). To avoid these problems, and also the problem of dating, researchers have assessed acidity changes in many of these pools by comparing preserved diatom samples collected in the early twentieth century and diatoms removed from the surface of old aquatic macrophyte herbarium material with recent samples that were collected in the same way.

Many of the Central European water bodies investigated (Figure 13; Tables 1 and 2) have a pH below 4.5 and contain only acidobiontic and acidophilous diatoms. Index B (Renberg & Hellberg, 1982), commonly used to infer past pH, does not work well in these situations. Therefore, a new index was developed based on weighted averaging of the pH optima of diatom taxa, and calibrated using a set of 97 pools and lakes of this region (Ter Braak & Van Dam, in press).

a. Denmark

Old and modern diatom assemblages from acidic Danish lakes have been analyzed and compared to determine historical pH changes (Table 2). The lakes and pools where no decrease or even an increase of DI pH is observed are more or less humic. Tormål, which is included here as a reference (or control), has the greatest alkalinity of the investigated sites (Ca^{2+}, 591 μeq/L; HCO_3, 414 μeq/L), and the increase of pH may be caused by hardening of the water, as has been observed in dune pools in The Netherlands by J.G.M. Roelofs (Catholic University, Nijmegen, pers. comm.). In general, DI pH decreases are less than 1 unit. Notable exceptions are Grønbakke, Grane Langsø, and Kongsø. Grønbakke is an ephemeral dune pond where the bottom is regularly exposed to the atmosphere. The water level of G. Langsø has risen considerably since the late 1970's and the water color has changed from clear to brown. This increased organic matter came from decomposition of drowned trees and other vegetation. The present low pH is possibly caused by humic acids. No changes in land use near Kongsø, close to G. Langsø, have occurred that might explain its present low pH. This small lake is situated in the open and the water is still very clear.

b. The Netherlands and Belgium

In The Netherlands and Belgium, most DI pH decreases are less than 1 unit (Tables 1 and 2). Pools for which no or very small changes of pH have been inferred are generally humic. Although the diatom inferred pH did not change in these pools, the species composition did change. In the old diatom samples, some taxa commonly found in humic acid water, e.g., Frustulia rhomboides var. saxonica (Rabh.) DeToni, Navicula subtilissima Cl., and Eunotia denticulata (Bréb.) Rabh., are more important than in new samples, which are from pools with a high mineral acidity and where E. exigua (Bréb. ex Kütz.) Rabh. is the most important taxon. The optimum of the latter species is 4.1 (Ter Braak & Van Dam, in press) and it is often the only diatom taxon in the most acidic

lakes; therefore, pH values cannot be inferred properly and acidification of the Dutch and Belgian sites is probably underestimated (Tables 1 and 2).

Deelense Was, where the pH has decreased 2.4 units, was used for washing sheep until the beginning of this century. The manure from the fleece caused an increase of alkalinity and pH of the pool. Relatively large changes of pH have also occurred in Achterste Goorven. The DI pH values in this pool decrease from the first to the third sampling stations. Historical research has shown that past higher pH levels were caused by inflow of agricultural drainage water at one end of this elongated pool during the nineteenth century (Dickman et al., 1987). Diatom analysis of a core from the central part of this pool (near station B) revealed that the pH increased from ca. 4.2 in 1810 to 6.2 in 1920 and then decreased to 4.5, partly due to termination of inflow of mesotrophic water by construction of a dam (Table 1). A similar pattern has been found in a core from the humic acid pool Kliplo (Table 1). This small peat bog was excavated about 1850 and subsequently eutrophied, presumably due to washing of sheep and/or foddering and catching of ducks. The pH increased to a peak of 6.2 in ca. 1910 and decreased to 5.0 in 1985. High pH values in the pool Gerritsfles in the early 20th century (Tables 1, 2) are due to sheep washing in the late 19th century (Van Dam et al., 1988).

Significant human activities have also occurred in and near some of the other pools listed in Table 1. These include soaking of timber (Kempesfles), bathing (Groot Huisven, Diepmeerven) and retting of flax (Onderste Rolven-E). Relatively large changes in DI pH ($\geq$ 0.7 units) have been observed for these pools. A part of the pH decline inferred for these other pools may have been caused by termination of traditional agricultural activities, but this cannot be determined with certainty because past human disturbance has not been documented.

c. West Germany, Austria, and Czechoslovakia

In West Germany, large DI pH decreases have been observed in Erdfallsee and Heideweiher (Table 2). Both water bodies received inflow of agricultural drainage water until a few decades ago. Comparison of the old and new samples from Plötschersee suggest an increase of 0.8 pH

units, which could have been caused by foddering of fishes. However, the coring results suggest a decrease of 0.5 pH units. Such differences, are occasionally found, but normally DI pH changes calculated from old and new samples and from cores are in good agreement (Arzet & Van Dam, 1986; Dickman et al., 1987; Van Dam, 1988b).

Some of the small lakes investigated in the Black Forest and the Bavarian Forest of southern Germany (e.g., Grosser and Kleiner Arbersee) have been used as reservoirs to increase the discharge of the outlet for wood-rafting purposes. This activity caused major fluctuations in water level and lake surface area until about the beginning of the present century. Forestry in the catchment areas has caused a shift from hardwood to softwood trees since the Middle Ages. The formerly mixed forest around Pinnsee was partially logged after World War II and reforested with conifers. Also, mining and smelting of heavy metals (Arbersees) and presence of local glass industry (Arbersees, Herrenwiesersee) may have changed the acidity status of some lakes in the past, including Teufelssee in Czechoslovakia (Arzet, 1987). Although some of these activities have left traces in the sediment profiles as small declines of DI pH, the acidification by acidic atmospheric deposition is clearly discernible as a rapid decrease of DI pH values since ca. 1950 (e.g., Grosser Arbersee; Figure 14) and shifts in chrysophyte species composition (Kleiner Arbersee; Hartmann and Steinberg, 1987). The recent acidification of some lakes in the northern Black Forest (Herrenwiesersee, Huzenbachersee, and Schurmsee) started between 1800 and 1850. Asterionella ralfsii decreased and Eunotia exigua increased in abundance during the period of acidification. (Steinberg et al., 1984; Arzet et al., 1986b; Arzet, 1987).

There is no evidence of a decrease in pH in the High Alpine lakes of Austria that have been investigated (Table 2; Arzet et al., 1986a).

IV. Discussion

Considerable high quality data now exist on which to make assessments concerning acidification trends in North America and Europe. As of 1988, sediment core DI pH data exist for at least 100 lakes on both continents (Tables 1 and 2). This number will probably double within the next two

years. The pH inference equations on which the DI pH data are calculated are based on 15 calibration data sets for North America and 10 for Europe, involving at least 500 and 300 lakes, respectively. The number of calibration lakes will also increase substantially in the next few years.

The ability of the existing predictive equations to infer pH is demonstrated in Table 1. With few exceptions, the surface sediment DI pH values agree within about 0.1 to 0.4 units of current measured pH.

A. Trends in pH and Extent of Acidification

The quantity and quality of diatom, chrysophyte, and other paleoecological data now available are sufficient to conclude that extensive regional lake acidification has occurred in low-alkalinity lakes of North America and Europe (Figure 15). However, it appears that sensitive water bodies in Europe are affected more by acidification than those in North America. At least 30% of the lakes studied in Europe had a DI pH decrease of > 0.6 pH units, whereas only one region in North America had more than 20% of its lakes in this category.

The lakes listed in Table 1 are not representative of all the lakes in their regions, but based on the studies done to date as summarized in this chapter, there is clear evidence of widespread acidification of low-alkalinity lakes in the Adirondack Mountains of New York, southeastern Canada, several regions in the United Kingdom, southern Sweden, Norway and Finland, The Netherlands, and northern Germany.

B. Causes of Lake Acidification

The primary cause of most recent lake acidification on both continents is atmospheric deposition of strong acids derived from combustion of fossil fuels, though watershed changes and probably natural long-term processes also play a role. The relative importance of these potential causal mechanisms can be evaluated using the hypothesis testing criteria described in section II.C.4. First, declines in pH in areas mentioned in Section IV. A. cannot be accounted for by natural

long-term processes (the rates of change are too fast) or watershed change, although the latter contributes, at least in some cases. Second, in the majority of cases, declines occurred after the onset of acidic deposition, and in many cases there is paleoecological evidence (e.g., Pb, V, polycyclic aromatic hydrocarbons, sphaerical carbonaceous particles) demonstrating that the fossil fuel derived inputs began before the pH declines. Third, all regions where pH declines are substantial (see above) receive relatively high atmospheric loading of strong acids.

The importance of watershed disturbance and change in affecting lake chemistry varies in kind and magnitude among the regions. In the northern United States and Canada, the most common disturbances are due to deforestation, forest fires, and blowdown. In Ontario, Canada, smelters have caused acidification of nearby lakes. In the western United States, factors such as increased deposition of windblown dust particles due to land use changes, ash falls from volcanoes (long term to the present), and localized concentrations of automobile emissions may be important. Recent acidification of some Florida lakes may be due partly or wholly to natural or anthropogenic lowering of the water table leading to reduced inflow of higher pH groundwater. In Europe, different types of recent anthropogenic land use changes are important, and have been occurring over a longer period of time. In the United Kingdom, afforestation rather than deforestation is of some importance. Decline in traditional agricultural practices, leading to regrowth of forest in former fields, has occurred in much of northern Europe and the United Kingdom; washing of sheep and foddering of ducks in The Netherlands have also contributed to change in pH. However, in none of the areas where considerable acidification has occurred can watershed change account for the inferred region-wide acidification.

C. Patterns of pH Trends

Although DI pH profiles exhibit a continuum of acidification trends (Figures 4 and 9), they can be classified arbitrarily into four basic patterns (Charles & Norton, 1986). Fluctuations in these patterns are caused by natural variability in diatom composition, watershed events, or inconsistencies introduced by pH inference methodologies. Two represent trends and two represent

no trend. The patterns are: (1) background pH greater than about 6.0 and no overall trend in pH, (2) relatively rapid decline in pH from the range of 5.7-6.0 to less than 5.0 (some pH declines start in pH ranges above 6.0), (3) pH about 4.8 to 5.2 throughout the entire profile, and no overall trend, and (4) decline from the range of pH 4.8-5.3 to as low as about 4.3.

The greatest changes in pH and sediment core diatom composition occur when DI pH decreases from a pH above approximately 5.5 to a pH below 5.5 (pattern 2). A smaller pH drop is observed for lakes with a pre-1850 DI pH of less than 5.5 (pattern 4). These patterns are consistent with the logarithmic nature of the pH scale and the response of sample lakewater pH to titration with strong acid in the laboratory. The pre-1850 water chemistry of lakes with patterns 1 and 2 and patterns 3 and 4 represent the dominant pre-1850 buffering systems in the studied regions: bicarbonate in the higher pH lakes, organic acids and Al species in the lower pH lakes. Declines in pH in the latter group (pattern 4) should be interpreted carefully. Though pH may have decreased only slightly, alkalinity may have decreased substantially. This is because the change in alkalinity per pH unit is greater in the pH range 4.5 to 5.0 than in the range 5.5 to 6.0. Also, an increase in Al concentration, which would be greater in the pH range 4.5 to 5.0, could have major impact on biota, much greater than the decline in pH alone. The effect on biota could be especially large if accompanied by a decrease in organic compounds that could complex with the Al.

D. Naturally Acidic Lakes

Lakes with pre-1850 DI pH less than 5.5 deserve special attention. First, they represent what most would consider "naturally" acidic lakes, and second, it is more difficult to explain their past chemistry and how it changed during the process of acidification. Many lakes of this type have been studied. For several regions (Tables 1 and 2), at least 40% of the lakes have pre-1850 DI pH < 5.5: Adirondacks, northern New England, northern Florida, Norway, Denmark, The Netherlands, Belgium, and Germany. There are two probable reasons for this high percentage. First, a region could have a high proportion of naturally acidic lakes due to its geologic setting

(e.g., Florida and The Netherlands) or second, because of conscious site selection criteria, only the more acidic lakes in a region were chosen for study, many of which turned out to be naturally acidic (e.g., New England).

Three important questions arise with respect to these naturally acidic lakes: (1) What was the chemistry of these lakes? (2) In particular, what were the dominant anions associated with the low pH? and (3) What specific chemical changes could have occurred in these lakes in response to increased deposition of strong acids, and how would those affect hydrogen ion concentration and alkalinity?

In regions with low-alkalinity waters that receive precipitation with pH greater than about 5.0, such as northeastern Minnesota and Wisconsin, western Ontario, Labrador (Barrie, 1984), and northwest Scotland (Barrett et al., 1983, 1987), clearwater lakes with pH less than 5.5 are rare (Wright, 1983). However, some lakes in these regions have pH less than 5.5. These lakes are often moderately to highly colored (platinum-cobalt units > 25) (Lillie & Mason, 1983; Gorham et al., 1985), associated with peatland vegetation, and only slightly influenced by groundwater input. Plants such as Sphagnum can remove cations from surrounding water (Clymo, 1963), and decay products of plants yield organic acids, which may or may not be colored (Vitt & Bayley, 1984). These organic acids can account for a significant portion of total anions in lake water and cause pH values to be less than 5.0 (Oliver et al., 1983). It seems probable, therefore, that organic acids were an important cause of acidic conditions in many lakes in acid sensitive areas before 1850 (Patrick et al., 1981; Arzet, 1987; Davis et al., 1985). Also, if the pH was about 5.0, Al concentrations may have been moderate at that time, although much of the Al would have been complexed with organic compounds (Driscoll et al., 1984).

As low pH (5.0 to 5.5) lakes have become more acidic, changes in Al and organic acid concentrations have probably been the most important factors affecting pH and alkalinity (e.g., Sullivan et al., 1989). The output of dissolved Al from these lakes' watersheds should have increased with increases in inputs of strong acids (Almer et al., 1978; Johnson et al., 1981; Driscoll et al., 1984), except perhaps for seepage lakes in outwash deposits. This is consistent with the fact that most of the lakes examined for this chapter that have current pH less than 5.5 have

concentrations of total Al greater than 100 to 200 μg/L; the value in some lakes is as high as 600 to 700 μg/L (e.g., National Research Council, 1986; Appendix Table E.3; Van Dam et al., 1981; Leuven, 1988; Van Dam, 1988a,b).

Increased Al concentration, at least in some Adirondack lakes, is suggested by the increase in percentages of diatom taxa such as Navicula tenuicephala (= ?Stauroneis gracillima) and Fragilaria acidobiontica toward the top of lake sediment cores (Charles et al., 1988). In the Adirondacks, these taxa are abundant only in lakes with pH less than 5.5 and total Al concentrations greater than 200 μeq/L (Charles, 1985, 1986). Also, computer modeling efforts based on current lake water chemistry and DI pH suggest that the Al concentration in Big Moose Lake increased as pH decreased (Charles, 1984).

Increased Al loading is important because it could lead to changes in alkalinity and reduced organic acids and water color. A relatively minor decline of pH in the range 4.5 to 5.5 can cause a major change in Al speciation and a substantial increase in total Al charge (Driscoll et al., 1984). Because Al acts as a buffer (Johannessen, 1980), increased concentrations can act to further minimize pH change as a result of increased acid loading.

Organic acids concentrations and water color can decrease as a result of decreasing pH and increasing Al concentrations. Support for these processes include theoretical analyses, laboratory studies, and observations that water color has decreased over time in lakes that have apparently acidified recently (Schofield, 1976; Almer et al., 1978; Dickson, 1980). Also, studies of limed lakes indicate that as alkalinity increases, color increases and concentration of Al decreases (Hultberg & Andersson, 1982; Yan, 1983; Wright, 1984). Van Dam (1987) studied the recovery of some moorland pools after the severe drought of 1976. The concentration of organic acids and water color increased, along with decreasing concentration of Al. At the same time, the proportional abundance of Eunotia exigua, characteristic of high mineral acids, decreased in favor of Frustulia rhomboides var. saxonica, which is commonly found in humic acid waters.

E. Taxonomic Composition

Many diatom taxa occur in all the regions discussed in this chapter, and in general, they all have similar ecological characteristics. The large and growing body of ecological data on diatoms in these regions provides additional support for the interpretations of a changing water chemistry inferred for any one region. The abundances of several taxa change consistently in sediment core profiles both within and among regions. Acidobiontic and acidophilic taxa that generally increase with increasing acidification in the Adirondacks, New England, eastern Canada, United Kingdom, Fennoscandia, and central Europe include Tabellaria binalis (except central Europe), Tabellaria quadriseptata, Frustulia rhomboides var. saxonica, Eunotia curvata (= E. lunaris), Navicula heimansii (= N. leptostriata), Navicula subtilissima types, and Eunotia exigua. In some regions, one or more taxa may be more important than others; for example Fragilaria acidobiontica and Navicula tenuicephala are important in currently acidic lakes with high Al in the Adirondacks, but the former has not been found in any European lakes. Eunotia exigua and other small Eunotia seem to be of much more importance in pools and lakes in The Netherlands and Germany than elsewhere.

Taxa that consistently decline with acidification in the above regions include Cyclotella kuetzingiana (primarily Europe), Cyclotella stelligera and Cyclotella comta (primarily North America and Fennoscandia), Achnanthes microcephala group, Anomoeoneis vitrea, and Fragilaria virescens. Many more taxa exhibit trends similar to those discussed above.

One of the most striking characteristics of diatom distributions both in modern assemblages and in sediment cores is the nearly universal absence of euplanktonic taxa where measured or DI pH is < 5.5 - 6.0. However, there are exceptions to this pattern. Asterionella ralfsii var. americana is apparently one of the few euplanktonic taxa found in lakes with pH < 5.5, at least in North America (e.g., Sweets, 1983). Also, recent studies of surface sediment diatom assemblages of lakes in the Killarney region of Ontario show that euplanktonic Cyclotella species are common in lakes with pH < 5.5. Lake morphometry may be an important reason for the difference in occurrence of euplanktonic diatoms in these lakes, compared with others (Taylor et al., 1987).

Chrysophyte assemblages also appear to change in a consistent manner with increasing acidification. In many lake regions, the preacidification chrysophyte flora are dominated by taxa such as Mallomonas crassisquama. However, as lakewater pH decreases, taxa such as Synura sphagnicola and S. echninulata often begin to increase. If acidification continues (e.g., to below pH of 5.0) even these species may be replaced by more acidobiontic forms such as M. hindonii, M. canina, and M. hamata. The often precipitous decline in M. crassisquama may be a useful marker of the onset of acidification, as is the decline in planktonic diatoms in many regions. Interestingly, changes in chrysophytes often predate shifts in diatoms by about 10 years. This may reflect a greater sensitivity of chrysophytes to subtle pH changes, perhaps because they are the often vernal blooming and planktonic.

F. Future Research

Despite the power of the diatom and chrysophyte-based paleoecological approach to inferring past acidification trends, improvements in methodology are possible. More research is needed on diatom and chrysophyte taxonomy, physiology, and ecology; in-lake transport and sedimentation processes; quantitative techniques for inferring water chemistry characteristics, including pH, alkalinity, dissolved organic carbon, and Al; statistical analysis of sediment core data; and lake selection criteria. For specific recommendations see Battarbee (1986), Charles & Norton (1986), and Davis & Smol (1986).

V. Conclusions

1. Analysis of sediment diatom and chrysophyte assemblages is the best technique currently available for inferring past lakewater pH trends and use of this approach is increasing rapidly.

2. Sediment core inferred pH data exist for at least 100 lakes in both North America and Europe. This number will approximately double in the next two years. The pH inference equations are based on at least 15 calibration data sets for North America and 10 for Europe, involving totals of at least 500 and 300 lakes, respectively.

3. Paleoecological studies indicate that recent acidification has been caused by acidic deposition in the Adirondack Mountains (New York), northern New England, Ontario, and Atlantic Provinces in North America; England, Scotland, Wales, Norway, Sweden, Finland, The Netherlands, and West Germany in Europe. Inferred pH decreases are commonly 0.5 to 1.5 pH units. Paleoecological and other data for several of the regions indicate combustion of fossil fuels as the source of acidic deposition. Watershed land use changes have caused some pH changes but cannot account for the pH changes on a regional scale.

4. No acidification trends were observed in regions currently receiving low deposition of strong acids (e.g., Rocky Mountains and Sierra Nevada in the western United States, northwest Norway, and northwest Scotland) and slight trends towards decreasing pH were observed in regions receiving moderately acidic deposition (Upper Midwest and Northern Florida, United States, and Atlantic Provinces of Canada).

5. The magnitude and extent of acidification of lakes appears to be greater in low-alkalinity regions of Europe than in North America. This is probably due to greater acid loading in Europe, regional differences in ability of watersheds to neutralize acids, and representation of lakes chosen for study.

6. Naturally acidic lakes (pre-1800 pH < 5.5) were apparently common in some regions, but rare in others.

7. Acidic deposition has decreased in some regions, but there is evidence of continuing acidification for some lakes.

8. Aluminum and organic acids are important in buffering pH changes, particularly in acidic waters.

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IX. FIGURE LEGENDS

1. Inferred lakewater pH calculated from multiple linear regression of diatom pH categories as measured surface water pH for 37 Adirondack lakes, and 95% confidence intervals for an individual prediction of pH from diatom data (from Charles & Smol, 1988).

2. Areas of North America where paleolimnological studies to infer past pH using diatom and chrysophytes are completed or in progress. Area boundaries include lakes in calibration data sets and those analyzed stratigraphically (see Table 1). Contours of precipitation pH are from Barrie and Hales (1984).

3. Lakes in the northeast United States and southeast Canada studied to infer historical diatom and chrysophyte pH. Lake names corresponding to map numbers, current pH, and inferred pH data are listed in Table 1.

4. Diatom inferred pH vs ^{210}Pb date for 10 Adirondack study lakes, calculated using multiple regression of Hustedt pH groups (Fig. 1, Charles & Smol, 1988). Error bars for DI pH values are the standard error of the regression (± 0.26 pH units). Error bars for ^{210}Pb dates are standard deviations (Binford, in press).

5. Diatom inferred pH for Speck Pond, Maine, calculated from multiple-regression analysis of taxa and pH groups and from log 10 index α (from Battarbee, 1984; Charles & Norton, 1986). Lead-210 dates have been added to published figure; data from R.B. Davis and S.A. Norton, University of Maine, Orono, pers. comm. CRS refers to Constant Rate of Supply model.

6. Chrysophyte taxa profiles for Lake Bonneville, Quebec (from Dixit et al., 1988d). See Table 1 and Fig. 3 for location and DI pH data. Mallomonas crassisquama generally occurs in higher pH lakes than does Synura echinulata. Reprinted by permission of Kluwer Academic Publishers.

7. Lochs and tarns in the United Kingdom studied to infer past lakewater pH by diatom analysis. Lake names corresponding to map numbers, current pH, and DI pH data are listed in Table 1. Lakes that have acidified recently are indicated by a black dot; those which have not changed, by an open circle.

8. Diatom taxa profiles for Llyn Llagi, Wales, showing changes in dominant species (from Battarbee et al., 1988). Synonyms for some taxa are: Navicula heimansii (= N. leptostriata), Eunotia lunaris (= E. curvata), E. veneris (= E. incisa).

9. Comparison of diatom inferred pH profiles for a series of sites in the United Kingdom with increasing lake water calcium values (Ca^{2+} as $\mu eq/L$) (from Battarbee et al., 1988).

10. Lakes in Norway, Sweden, and Finland, studied to infer past lakewater pH by diatom analysis. Lake names corresponding to map numbers, current pH and inferred pH data are listed in Table 1. Lakes that have acidified recently are indicated by black dots; those which have not changed, by an open circle. Lake 42 is in the same location as lake 36.

11. Diatom pH spectra in a sediment core from Holmvatn, S. Norway (left) and diatom inferred pH and TOC (right). In the 1970s, the average measured pH was 4.7 and the average measured TOC was 2.2 ppm. Based on diatom studies, Holmvatn was a naturally acidic, moderately humic lake that has become more acidic and much less humic since ca. 1930s. The absence of watershed changes correlated with the acidification suggests that acid

deposition was the cause (from Davis, 1987). AcB = acidobiontic; Ac = acidophilic; IND = "indifferent"; Al = alkaliphilic.

12. Stratigraphy of selected diatom taxa, Hustedt's pH categories, and inferred pH using index B, for a core from Gårdsjön, SW Sweden. The development in Gårdsjön is typical for a large number of Swedish lakes and shows a severe recent acidification reflected by a marked change of the subfossil diatom record of the sediments (from Renberg & Hellberg, 1982; Renberg & Wallin, 1985).

13. Lakes and pools in Western Europe studied to infer past lakewater pH by diatom analysis. Lake names corresponding to map numbers, current pH and inferred pH data are listed in Table's 1 and 2. [Currently 2 maps, but these will be combined]

14. Relative frequency of selected diatom taxa and inferred pH based on Index α and Index B for Grosser Arbersee (from Arzet et al., 1986). Eunotia veneris is a synonym of E. incisa.

15. Decrease of DI pH in 10 different regions. The length of each graph is 100%. The number of localities in each region is shown at right. All lakes listed in Table 1 are included, except those artificially acidified. Five classes of decrease of pH from pre-industrial times until ca 1980-1985 are distinguished. The lakes were not selected randomly, and do not represent all lakes in a region. They show results only for a particular group of lakes studied to date.

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Table 1. Summary of the extent and timing of pH changes for lakes in North America and Europe. North American regions are ordered west to east; European regions west to east. See Figs. 3, 7, 10, and 13 for locations of lakes. References are as follows: (1) Del Prete & Schofield, 1981; (2) Charles, unpublished data; (3) Davis et al., 1987; (5) Charles et al., 1989 ; (6) Rhodes, 1985; (7) Norton et al., 1981; (8) R.B. Davis & D.S. Anderson, pers. comm.; (9) Davis et al., 1983 (10) Ford, 1986; (11) R.B. Davis, M.C. Whiting, & D.S. Anderson, pers. comm.; (12) Winkler, 1985, 1988; (13) Fortescue, 1984; (14) Dickman et al., 1983; (15) Delorme et al., 1985; (16) Dixit, 1986b; Dixit & Evans, 1986; Dixit et al. 1987a; (17) McKee et al., 1987; (18) Hudon et al., 1986; (19) Dixit et al., 1987b, 1988c,d; S.Dixit, Queen's University, pers. comm.; (20) Elner & Ray, 1987; (21) Scruton & Elner, 1986; Scruton et al., 1987; (22) Scruton et al., 1987; (23) Davidson, 1984; Dickman et al., 1988; (24) Kingston et al., 1989; (25) Kingston & Kreis, 1987; (26) Sweets et al., 1989; (27) Baron et al., 1986; (28) Stuart, 1984; Norton & Kahl, 1987; Beeson, 1988; (29) Holmes, 1986; (30) M.C. Whiting & D.R. Whitehead, pers. comm.; (31) Battarbee et al., 1988; (32) Berge, 1975; (33) Berge, 1979; (34) Davis & Berge, 1980; (35) Davis et al., 1983; (36) Davis et al., 1980; (37) Davis, 1987; (38) Berge, pers. comm.; (39) Berge & Birks, pers. comm.; (40) Miller, 1973; (41) Renberg & Hellberg, 1982; (42) Renberg, unpublished; (43) Tolonen & Jaakkola, 1983; (44) Tolonen et al., 1986; (45) Simola et al., 1985; (46) Tolonen & Suksi, pers. comm.; (47) Liukkonen, pers. comm.; (48) Turkia, pers. comm.; (49) Van Dam et al., 1988; (50) Dickman et al., 1987; (51) Arzet et al., 1986a; (52) Arzet, 1987; (53) Steinberg et al., 1984; (54) Arzet et al., 1986b. E.L. = Elk L., VT = Vermont, ME = Maine, NH = New Hampshire, MA = Massachusetts, MI = Michigan, MN = Minnesota, WI = Wisconsin, CO = Colorado, Wy = Wyoming, NB = New Brunswick, NS = Nova Scotia, nd = no data, ns = not significant (pH decrease $\leq$ 0.2 units) na = not applicable, du = ^{210}Pb dating uncertain or core not dated.

Map num.	Lake	Year of core	Current lakewater pH	Surface sediment inferred pH	Inferred pH (pre-1850)	Inferred pH change[a]	Approximate date of first pH change	Primary cause of pH change	Ref.
NORTH AMERICA									
Fig. 3									
(a)	**USA, Adirondack Park, NY**								
1	Honnedaga L.	1976	4.7-4.8	5.2	6.1	-1.1	na	A	1, 2
2	Seventh L.	1976	6.4-7.0	6.5	6.6	ns	na	na	1

3	Woodhull L.	1976	5.1-5.3	6.0	6.1	ns	na	na	1
4	Panther L.	1980	7.2	6.9	7.5	-0.6	post-1960	A	3
5	Sagamore L.	1978	6.0	6.3	6.8	-0.5	1940s	A	3
6	Woods L.	1980	4.9	4.8	5.2	-0.4	1930s	A	3
7	Barnes L.	1983	4.7	4.7	5.2	-0.5	post-1900	A	4
8	Bear P.	1983	5.0	5.5	5.9	-0.4	1920-1940	A	4
9	Big Moose L.	1982	4.6-5.1	5.0	6.0	-1.0	1920-1940	A	4, 5
10	Clear P. (E.L.)	1982	7.0	7.0	7.0	ns	na	na	4
11	Deep L.	1983	4.7	4.8	5.4	-0.6	1940-1950	A	4
12	L. Arnold	1982	4.8	4.7	5.1	-0.4	du	A	4
13	Little Echo P.	1982	4.8	4.5	4.6	ns	na	na	4
14	Merriam L.	1982	4.9	5.1	5.4	-0.3	1910-1930	A	4
15	Queer L.	1983	5.3	5.4	6.4	-1.0	1930-1950	A	4
16	Upper Wallface P.	1983	5.0	4.9	5.3	-0.4	1910-1930	A	4
17	West P.	1982	5.5	5.7	5.6	ns	na	na	4
18	Windfall P.	1982	6.5	6.9	6.8	ns	na	na	4
19	Holmes L.	1984	5.0(1980)	limed	5.3	?	na	na	6
	(b) USA, New England								
20	Branch P. VT	1980	4.7	4.5	4.8	-0.3	1930	A	7, 8
21	Cone P. NH	1980	4.5-4.8	4.6-4.7	4.5-4.8	ns	na	na	10
22	E. Chairback P. ME	1979	5.2	5.0-5.1	4.8-4.9	+0.2	1920-1930	A	9
23	Haystack P. VT	1983	4.8	5.0	5.3	-0.3	1920	A	11
24	Klondike P. ME	1978	6.2	5.5	5.7	ns	na	na	9
25	Ledge P. ME	1978	5.6	4.4-4.7	5.2	-0.6	1880s	A	9
26	Little Long P. ME	1983	5.6	5.8	6.0	-0.2	1950	A	11
27	Mountain P. ME	1979	6.1	5.0	5.1	ns	na	na	7, 8
28	Mud P. ME	1983	4.5	4.7	5.2	-0.5	1950	A	11
29	Solitude P. NH	1979	4.8	5.0	4.9-5.2	ns	na	na	9

30	Speck P. ME	1978	5.2	4.9	5.1	ns	na	na	9
31	Tumbledown P. ME	1978	5.2	5.1	5.0	ns	na	na	9
32	Unnamed P. ME	1979	4.8	5.1	5.1	ns	na	na	9
33	Duck P. MA	1983	4.6	5.0	5.2	-0.2	du	?	12
	(c) Canada, Ontario								
34	Lake B	1981	5.2	4.8	6.4-6.6	-1.6	1955	S	13, 14
35	Lake CS	1981	5.2	6.4	7.2	-1.0	1955	S	14
36	Batschawana L.	1981	6.1	6.2	6.0	ns	na	na	15
37	Clearwater L.	1984	4.5	4.3	6.3	-2.0	1930	S	16
38	Hannah L.	1984	4.3(1970)	4.6(1970)	6.0	-1.4	1880	S	16
39	Lohi L.	1984	4.7	nd	nd	na	na	na	16
40	Quirke L.	1984	5.5-6.4	5.5	6.3	-0.8	1955-1959	S	17
	(d) Canada, Quebec								
41	C-22	1983	5.8	5.4	5.5	ns	na	na	18
42	Key L.	1983	5.5	5.2	5.3	ns	na	na	18
43	Blais L.	1986	7.1	7.2	7.3	ns	du	na	19
44	Bonneville L.	1986	5.4	5.4	5.6	-0.2	1940	A	19
45	Chevreuil L.	1986	7.0	7.1	7.0	ns	na	na	19
46	Chomeut	1986	6.7	6.9	7.0	ns	na	na	19
47	David L.	1986	7.3	7.1	7.0	ns	du	na	19
48	Kidney L.	1986	6.8	6.8	6.8	ns	na	na	19
49	Lagou L.	1986	5.8	5.6	5.7	ns	na	na	19
50	Lemaine L.	1986	6.0	6.2	6.1	ns	na	na	19
51	Nolette L.	1986	6.5	6.4	6.5	ns	na	na	19
52	Thomas L.	1986	6.8	6.4	6.5	ns	na	na	19
53	Truite Rouge L.	1986	6.1	5.7	5.8	ns	1943	A	19
	(e) Canada, New Brunswick and Nova Scotia								
54	Big Indian L. NS	1981	nd	5.3	6.1	-0.8	post-1939	A	20

55	Emigrant L. NB	1981?	6.1	6.1	6.5	-0.4	pre-1948	A	20
56	Kinsac L. NS	1981	5.5	6.1	6.3	-0.2	post-1917	A	20
57	Lilly L. NB	1981?	5.6	5.7	6.2	-0.5	du	A	20
58	Round L. NS	1981	4.6	6.2	6.2	ns	na	na	20
59	St. Patricks L. NB	1981?	6.7	6.8	6.8	ns	na	na	20
60	Tomoowa L. NB	1981?	5.3	6.4	6.4	ns	na	na	20
61	Tupper L. NS	1981	nd	nd	nd	nd	na	na	20
62	8	1984	5.3	5.3	5.3-5.6	-0.2-0.3	du	na	21
63	12	1984	5.1	5.1	5.3-5.4	-0.2-0.3	du	A?	21
64	204	1984	5.1	5.2	5.5	-0.3	du	A?	21
65	219	1984	5.6	5.5	5.5	ns	na	na	21
66	660	1984	5.5	5.6	5.7	ns	na	na	21
67	817	1984	5.2	5.3	5.7	-0.4	1930	A?	21
	(f) Canada, Newfoundland								
68	Aides P.	1984	6.0	6.4	6.0	+0.4	1930-1940	na	22
69	Lake # 660	1984	5.5	5.5	5.7	-0.2	1946		21
70	Stephensons P. (206)	1984	5.6	5.9	6.0	ns	na	na	22
	(g) Canada, NW Ontario, Experimental Lakes Area								
71	L. 223	1984	5.3-5.5	5.4	6.7-7.0	-1.4	1976?	E	23
	(h) USA, Northern Minnesota, Wisconsin and Michigan								
72	Andrus L. MI	1984	5.6	6.0	6.0	ns	na	na	24
73	Brown L. WI	1984	5.7	5.9	6.4	-0.3	1900	?	24
74	Camp 12 L. WI	1984	5.4	5.6	5.6	ns	na	na	24
75	Denton L. WI	1984	5.2	5.5	5.7	-0.3	1870	?	24
76	Dunnigan L. MN	1984	6.7	6.4	6.6	ns	na	na	24
77	Hustler L. MN	1984	6.8	6.9	6.6	+0.3	na	na	24
78	McNearney L. MI	1984	4.4	4.9	4.9	ns	na	na	24
79	Nels L. MN	1984	6.4	6.5	6.6	ns	na	na	24

80	Otto Mielke L. WI	1984	5.3	5.8	5.4	+0.4	na	na	24
81	Bastile L. WI	1986	5.0	5.6	5.6	ns	na	na	25
82	Hillis L. WI	1986	5.6	5.5	5.8	-0.3	du	A?	25
83	McGrath L. WI	1986	5.2	5.4	5.6	-0.2	du	na	25
84	Morgan L. WI	1986	4.6	5.4	5.5	ns	du	na	25
85	Sugar Camp L. WI	1986	5.4	5.7	5.0	ns	na	na	25
	(i) USA, N. Florida								
	Fore L.	1983	5.1	5.1	5.0	ns	na	na	26
	L. Barco	1983	4.5	4.2	4.9	-0.7	1958	A?,F?	26
	L. Lou	1983	6.3	6.8	6.8	ns	na	na	26
	L. Mary	1983	4.6	4.8	4.9	ns	na	na	26
	L. Suggs	1983	4.9	5.4	6.3	-0.9	late 1800s	A?,F?	26
	Mirrow L.	1983	4.9	5.0	5.1	ns	na	na	26
	(j) USA, Rocky Mountains								
	Emerald L. CO	1982	6.7	6.7	6.8	ns	na	na	27
	L. Haiyaha CO	1982	6.3	6.8	6.7	ns	na	na	27
	L. Husted CO	1982	6.8	6.7	6.5	ns	na	na	27
	L. Louise CO	1982	6.8	6.9	6.8	ns	na	na	27
	Black Joe L. WY	1984	6.6	6.8	6.8	ns	na	na	28
	Deep L. WY	1984	nd	6.7	6.7	ns	na	na	28
	Hobbs L. WY	1984	6.4	6.8	6.8	ns	na	na	28
	(k)USA, California, Sierra Nevada								
	Emerald L.	1986	6.3	6.6	6.4	+0.2	na	na	29, 30
	Eastern Brook L.	1986	7.1	7.1	6.9	+0.2	na	na	30
	Harriet L.	1986	6.5	6.4	6.4	ns	na	na	30
	Lake 45	1986	5.2	5.7	5.9	-0.2	na	na	30

EUROPE

Fig. 7

	(l) Wales								
1	Llyn Hir	1984	4.8[d]	nd	6.1	-1.3[e]	1850	A	31
2	Llyn Gynon	1985	5.2	5.5	6.0	-0.5	1945	A	31
3	Llyn y Bi	1985	4.9	4.7	6.0	-1.3	1900	A	31
4	Llyn Dulyn	1985	4.9	4.7	6.0	-1.3	1850	A	31
5	Llyn Eiddew Bach	1985	4.8	nd	6.6	-1.8	1940?	A	31
6	Llyn cwm Mynach	1985	5.6	5.6	6.1	-0.5	1850	A	31
7	Llyn Llagi	1985	4.9	4.6	6.1	-1.5	1850	A	31
	(m) Scotland								
8	Loch Enoch	1982	4.5	4.4	5.3	-0.9	1840	A	31
9	Loch Valley	1981	4.7	4.6	5.6	-1.0	1860	A	31
10	Round L. of Glenhead	1981	4.7	4.9	5.9	-1.0	1860	A	31
11	Loch Dee	1980	5.3	5.5	6.2	-0.7	1890	A	31
12	Loch Grannoch	1980	4.7	4.7	5.7	-1.0	1930	A	31
13	Loch Fleet	1985	4.5	4.6	5.8	-1.2	1975	A,R	31
14	Loch Skerrow	1980	5.3	5.8	5.9	ns	na	na	31
15	Loch Urr	1984	6.8	6.8	6.5	ns	na	na	31
16	Loch Tanna	1986	5.0	4.6	5.0	-0.4	1850	A	31
17	Loch Tinker	1985	5.7	6.0	6.4	-0.4	1850	A	31
18	Loch Chon	1986	5.4	5.5	6.2	-0.7	1900[c]	A,R	31
19	Loch Laidon	1985	5.4	5.3	5.8	-0.5	1860	A	31
20	Lochnagar	1986	5.0	4.8	5.7	-0.9	1850	A	31
21	Dubh Loch	1986	5.3	5.2	5.7[b]	-0.5	du	A	31
22	Loch nan Eun	1986	5.0	4.8	6.0[b]	-1.2	du	A	31
23	Lochan Uaine	1986	5.8	5.7	5.8	ns	na	na	31
24	Lochan Dubh	1986	5.6	5.2	5.5	-0.3	du	A	31

	(n) England								
25	Scoat Tarn	1984	5.0	4.6	6.0	-1.4	1850	A	31
26	Low Tarn	1984	5.0	4.6	6.0[b]	-1.4	du	A	31
27	Greendale Tarn	1984	5.2	4.9	6.0	-1.1	1900	A	31
28	Burnmoor Tarn	1979	6.4	6.2	6.3	ns	na	na	31
Fig. 10									
	(o) Norway								
1	Langtjern	1975	4.7-5.2	<5.5	4.3-6.2	ns	na	na	32
2	Övre Målmesvatn	1977	4.5	<5.2	4.4-6.5	-0.5	1930	A	33
3	Blåvatn	1978	5.1	5.1	5.2	-0.1	1930	A	34,35
4	Hovvatn	1978	4.4	3.9-4.4	4.8-5.4	-0.7	1918	A	34,35
5	Brårvatn	1979	5.2	5.2-5.3	5.6-6.3	-0.7	1900	A	35,36,37
6	Botnavatn	1978	5.7	5.7	5.9	-0.2	1920	A	35
7	Dorsvatn	1978	5.0	5.0	5.0	ns	na	na	35
8	Grönlivatn	1978	6.5	6.5	6.5	ns	na	na	35
9	Holmvatn	1978	4.7	4.6	4.9-5.2	-0.5	1930	A	35,37
10	Nedre Målmesvatn	1979	4.6	4.4-4.5	4.9-5.3	-0.6	1890	A	35
11	Oppljosvatn	1979	5.8	5.8	5.8	ns	na	na	35
12	Gulspettvatn	1985	4.9	4.8	5.6	-0.8	du	A	38
13	Röyrtjörna	1987	6.5	6.0	6.0	ns	na	na	38
14	Verevatn	1986	4.4	4.4	5.1	-0.7	du	A	38
15	Holetjern	1986	4.6	4.6	5.1	-0.5	du	A	39
16	Ljosvatn	1986	4.5	4.6	5.2	-0.6	du	A	39
17	Skomakarvatn	1987	4.6	4.5	5.1	-0.6	du	A	39
	(p) Sweden								
18	Stora Skarsjön	1971	4.5	4.5	6.0	-1.5	du	A	40
19	Gårdsjön	1979	4.6	4.5	6.0	-1.5	1950	A	41
20	Härsvatten	1979	4.4	4.2	5.9	-1.7	du	A	41

21	Lysevatten	1979	5.9	5.2[f]	6.1	-0.9[f]	du	A	41
22	Lilla Öresjön	1986	4.5	4.6	6.1	-1.5	du	A	42
	(q) Finland								
23	Hauklampi	1982	4.8-4.9	5.1-5.4	6.0-6.4	-1.0	1962	A	43
24	Häklājärvi	1982	4.8-5.2	5.6	6.3	-0.7	du	A	43
25	Orajärvi	1982	4.7-4.8	4.9	6.3	-1.4	1930	A	43
26	Hirvilampi	1983	5.6	5.5	6.0-6.4	-0.7	1960	A	44
27	Matalajärvi	1983	4.7	4.9	5.8-6.2	-1.0	1950	A	44
28	Munajärvi	1983	4.7	5.2	5.5-6.0	-0.6	1950	A	44
29	Pappilanlampi	1966	5.3	5.0-5.5	5.0-5.5	ns	na	na	44
30	Työtjärvi	1975	5.1-6.7	6.6-7.0	6.6-7.0	ns	na	na	44
31	Valkeislampi	1983	5.1	5.2	5.2	ns	na	na	44
32	Valkjärvi	1983	5.1	5.5-5.7	6.1-6.4	-0.7	1930	A	44
33	Vitsjön I	1979	5.9	5.9-6.4	6.8	-1.0	du	W	44
34	Vitsjön II	1983	4.3	5.6	6.0-6.2	-0.5	1930	A	44
35	Vuorilampi	1983	5.9	5.3-5.5	6.3-6.5	-1.0	1960	W	44
36	Iso-Hanhijärvi	1984	4.9	5.0	5.0-5.5	-0.3[g]	du	A[g]	45
37	Kakkisenlampi	1984	4.8	5.0	4.5-5.1	ns	na	na	45
38	Kangasjärvi	1984	5.3	5.2	5.2-5.5	-0.3[g]	1960	A[g]	45
39	Kiiskilampi	1984	4.8	4.3	4.7	-0.4	du	A	45
40	Pieni-Kalliojärvi	1984	5.2	5.0	nd	-	na	na	45
41	Pieni-Löytönen	1984	5.1	6.0	6.5-7.0	-0.8[g]	du	A[g]	45
42	Siikajärvi	1984	5.3	5.1	5.9	-0.8	1960	A	45
43	Sonnanen	1984	6.5	6.5	6.5	ns	na	na	45
44	Suo-Valkeinen	1984	4.9	4.7	5.0	-0.3	1965	A	45
45	Valkealampi	1984	4.9[h]	4.7	4.5-4.8	ns	na	na	45
46	Iso-Tiilijärvi	1983	5.1	5.2-5.7	5.6-6.0	-0.2	du	A	46
47	Kankareenjärvi	1984	5.4	5.4-5.9	5.7-6.2	-0.3	1965	A	46

48	Koukjärvi	1984	5.6	5.5-6.0	5.7-6.4	-0.3	1965	A	46
									47
49-72	Work in progress; See Fig. 10 for pH change								48
73-78	Work in progress; See Fig. 10 for pH change								
Fig. 13									
	(r) The Netherlands								
1	Kliplo	1985	5.3	5.0	4.5	-1.2[i,g]	1910	A,W	49
2	Gerritsfles	1985	4.3	4.2	4.4	-1.7[i,g]	1900	A,W	49
3	Achterste Goorven	1985	3.9	4.5	4.2	-1.7[i,g]	1920	A,W	50
	(s) West Germany								
4	Pinnsee	1983	-	4.7	6.0	-1.3	1930	A,W	51
5	Plötschersee	1984	5.8	6.0	6.5	-0.5	after 1900	A,W?	52
6	Grosser Bullensee	1983	3.7	3.7	4.5	-0.8	after 1930	A,W	52
7	Kleiner Bullensee	1983	3.7	3.7	4.5	-0.8	after 1930	A,W	52
8	Oderteich	1984	4.5	4.8	5.1	-0.3	du	A,W,F	52
9	Herrenwiesersee	1983	3.6-4.1	3.7-4.3	4.5	-0.2-0.7	1880	A,W,F	51
10	Huzenbachersee	1984	4.3	4.3	5.0	-0.7	c. 1850	A,W,F	52
11	Schurmsee	1984	4.1	4.0	4.7	-0.7	c. 1850	A,W,F	52
12	Wildsee (Ruhestein)	1984	4.3	4.4	4.4	ns	na	na	52
13	Glaswaldsee	1984	4.3	4.8	4.8	ns	na	na	52
14	Feldsee	1983	6.3	6.0	6.1	ns	na	na	52
15	Grosser Arbersee	1983	4.5-5.1	4.7	5.5	-0.8	1960	A,S,F	51
16	Kleiner Arbersee	1981	4.5-5.1	4.7	5.5	-0.8	1930	A,F	51
17	Rachelsee	1983	4.3-4.9	4.4-4.6	5.1-5.2	-0.5	1950	A,S,F	53
	(t) Austria								
18	Mutterbergsee	1985	5.2	5.2	4.8	+0.4	du	na	52
19	Oberer Plenderlesee	1984	7.0	7.3	6.9-7.1	+0.3	du	na	54
20	Schwarzsee ob Solden	1984	5.2-5.4	5.1	5.1-5.5	ns	na	na	54
21	Goasselesee	1984	6.2	6.0	6.0-6.1	ns	na	na	54

22	Gippersee	1984	6.2	6.9	6.7-6.8	ns	na	na	54
	(u)Czechoslovakia								
23	Teufelssee	1986	4.3	4.3	4.8	-0.5	after 1900	A,F,W	52

Primary cause of pH change (other factors may also contribute): A = Acid deposition (long-range transport); S = local acid deposition source; L = Liming; E = experimental acidification; W = Watershed disturbance (e.g., logging, fire); H = Sheep washing; R = Reforestation?; F = Water level changes

a Surface sediment minus pre-1850 DI pH.
b Calculated from sample at base of the core.
c Main change after reforestation (post-1970).
d Pre-liming data.
e Calculated using modern measured pH.
f Affected by liming in 1974.
g Reacidified after cultural eutrophication.
h Before liming.
i Maximal DI pH (1900-1920) minus current diatom inferred pH.

Table 2. Changes in diatom inferred pH between old and new samples from pools and lakes in Western and Central Europe. When more than one sample is available, the number of samples is given between parentheses and the values are medians. Inferred pH values are calculated by weighted averaging (Ter Braak & Van Dam, 1988). Diatom counts for localities 19, 23, 33, 34 and 37 from Van Dam et al. (1981), for localities 22, 25 and 36 from Van Dam (1988), other diatom counts original. New pH measurements were done simultaneously with diatom sampling and taken from various sources (Van Dam, in prep.). Inferred pH changes less than 0.3 units are not considered to be significant, unless coinciding with important changes in the relative abundance of the acidobiontic diatom Eunotia exigua. Apparent causes of pH change: A = acid deposition (long-range transport), s = local acid deposition source, W = watershed disturbance (e.g. logging, sheep washing), F = changes of water level).

Nr	Name	Depth (m)	Area (ha)	Altitude (m)	Old sampling (diatoms)	Inferred old pH	New sampling (diatoms)	Inferred new pH	Inferred pH change	New sampling (pH meas.)	New pH (meas.)	Apparent cause of pH change
Denmark												
1	Lille Helmiskaer-W.	0.1	0.69	9	1945	6.9	1982	5.6	-1.3	1982	5.1	A
2	Birkemose	0.5	2.69	7	1945	6.0	1982	5.0	-1.0	1982	4.7	A
3	Hosse	6.0	3.63	70	1941	5.1	1982	4.3	-0.8	1982	4.2	A
4	Lille Økse	5.5	0.34	62	1943	4.3	1982	4.2	ns	1982	3.7	A
5	Sokland	1.0?	11.50	16	1959	5.0	1982	5.1	ns	1977-82(23)	5.2	ns
6	Tornø	0.5	7.71	16	1950	6.2	1982	6.6	+0.4	1982	7.3	ns
7	Praestekaer 2	1.0?	1.34	16	1967	5.1	1982	5.0	-0.1	1977-82(23)	5.0	A
8	Nykaer	1.0	13.00	16	1950	4.5	1982	5.2	+0.7	1977-82(27)	4.6	ns
9	Grønbakke	0.1	3.38	5	1943	7.0	1982	5.1	-1.9	1982	4.9	A
10	Hundsø	7.0	1.81	75	1944	4.9	1982	4.8	-0.1	1977-82(27)	4.4	A
11	Raevsø	8.0	5.00	74	1949	6.7	1977-82(2)	6.3	-0.4	1977-82(27)	6.0	A
12	Grane Langsø	11.5	11.40	74	1949	6.5	1977-82(2)	4.9	-1.6	1977-82(27)	5.3	A
13	Kalgaard Sø	11.0	10.50	74	1944	6.5	1982	6.5	ns	1976-82(37)	6.7	ns
14	Kongsø	9.3	3.00	74	1949	7.2	1982	4.8	-2.4	1977-82(27)	5.0	A
15	Lille Gribsø	6.0	0.39	50	1943	4.7	1982	4.8	ns	1982	5.0	A
16	Njortesøle	3.5	0.05	10	1943(2)	5.1	1982(2)	5.5	+0.4	1982	6.1	ns
17	Sorte Sø	4.8	1.10	66	1947	5.7	1982	5.2	-0.5	1982	4.8	A
The Netherlands												
18	Van Nunenplak	0.8	0.69	4	1963	5.6	1983	5.0	-0.6	1979-83(16)	4.5	A
19	Reeënveen	1.0?	0.19	15	1929	4.2	1978	4.2	ns	1978	4.3	ns
20	Diepveen	1.2	0.81	10	1924	4.9	1978-86(3)	4.2	-0.7	1978-86(3)	4.5	A
21	Poort 2	0.5	0.01	10	1924(2)	4.3	1978-86(4)	4.2	ns	1978-86(3)	4.5	ns
22	Kliplo	1.1	0.62	13	1924-29(2)	5.0	1972-84(11)	4.6	-0.4	1970-84(34)	5.3	A
23	Schurenberg	1.3	1.00	13	1924	4.5	1978	4.6	+0.1	1978	3.8	A
24	Pool in Echtenerzand	0.5	0.25	10	1933	4.4	1978-86(3)	4.1	-0.3	1978-82(3)	3.7	A
25	Gerritsfles	1.2	6.78	40	1916-18(7)	4.8	1964-84(21)	4.1	-0.7	1974-84(18)	4.3	A,W
26	Kempesfles	0.5	0.28	25	1918	4.8	1973-86(4)	4.1	-0.7	1973-86(4)	3.6	A,W
27	Doelense Waa	0.6	0.50	45	1917	6.4	1983	4.2	-2.4	1983(7)	4.1	A,W
28	Bloempjesven	1.0	0.63	17	1918	4.4	1982	4.1	-0.3	1982	3.8	A
29	Ganzenven	0.2	0.28	7	1950	4.1	1982	4.1	0.0	1982	4.3	A
30	Goudbergven	1.0	1.45	13	1929	4.2	1982	4.0	-0.2	1982	3.7	A
31	Zwarte Goor	0.4	5.25	12	1929	4.4	1982	4.0	-0.4	1982	3.7	A,W
32	Schaapsven	1.0	1.56	11	1919-25(2)	4.6	1975-82(3)	4.1	-0.5	1975-86(5)	4.5	A,W
33	Galgeven	3.6	11.30	11	1919	4.9	1978	4.2	-0.7	1978	4.0	A
34	Diaconieven	1.6	1.45	10	1916	4.4	1978	4.0	-0.4	1978	3.9	A
35	Brandven	1.0	1.40	10	1926	4.3	1978	4.0	-0.3	1978	3.8	A
36A	Achterste Goorven A	1.8	2.35	10	1919-29(3)	6.8	1978-84(11)	4.3	-2.5	1975-84(18)	4.1	A,W
36B	Achterste Goorven B	1.8	2.35	10	1925-29(5)	5.8	1975-84(12)	4.1	-1.7	1975-84(20)	3.9	A,W
36E	Achterste Goorven E	1.3	1.10	10	1919-28(12)	4.7	1975-28(12)	4.0	-0.7	1978-84(19)	3.9	A,W
37	Klein Adervan	0.6	0.72	9	1921(2)	4.5	1978	4.0	-0.5	1978	4.2	A
38	Middelste Wolfsputven	1.0	0.09	8	1919	4.4	1978-82(3)	4.3	-0.1	1978-86(5)	4.6	A
39	Tongbreven-W.	1.7	3.36	8	1929(2)	4.4	1984	4.6	ns	1984	4.9	ns
40	Groot Huisven	1.5	2.80	25	1932	5.0	1976-86(9)	4.2	-0.8	1978-86(25)	4.7	A
41	Diepeerven	1.0	0.31	20	1952	4.8	1982	4.0	-0.8	1981-82(2)	3.5	A,W
42	Pikmeeuwenwater-SE	0.6	0.40	25	1952	4.5	1982	4.4	-0.1	1982	4.2	A
43	Ravenven-NW	1.0	2.13	30	1957	4.4	1982	4.0	-0.4	1982	3.7	A
44	Rouwkuilenven	0.5	0.25	43	1956	4.3	1983	4.4	+0.1	1982-83(45)	4.9	A
45	Onderste Holven-E					5.4	1983	4.5	-0.9	1983	4.6	A
Belgium												
46	Pool W. of Zwartwater	0.5	0.22	29	1935	4.5	1982(2)	4.1	-0.4	1975-82(18)	3.9	A
47	Pool N.W. of Zwartwater	0.5	0.10	29	1935	4.5	1982	4.1	-0.6	1975-82(17)	3.7	A
48	2 pools in Fagne d'Ardenne	2.0	0.005-.01	650	1952-53(2)	4.3	1982(2)	4.3	0.0	1982(2)	3.9	A,W
49	2 pools in Fagne de Massebotté	0.2	0.02	590	1949(2)	4.3	1982(2)	4.6	+0.3	1982	4.6	ns
50	Pool in Grande Fagne	0.2	0.003	560	1952	4.5	1982	4.4	ns	1982(2)	4.8	ns
East Germany												
51	Filzteichersee	13.3	8.68	42	1924	5.2	1983	6.0	+0.8	1983(1)	5.8	ns
52	Grosser Bullensee	11.0	9.80	32	1928	4.4	1983	4.0	-0.4	1983(20)	3.8	A
53	Erdfallsee	11.3	4.50	43	1928	7.1	1983	5.9	-1.2	1978-83(14)	6.1	A,W
54	Reideweiher	1.0	1.92	44	1928	6.1	1983	4.6	-1.5	1983	4.9	A,W
55	Weinfelder Maar	52.0	15.90	484	1942	7.6	1983	7.4	ns	1977-83(3)	7.3	ns
56	Wildsee (Kaltenbronn)	3.0	2.30	908	1961	4.1	1983(2)	4.1	0.0	1983(2)	3.6	A, F
57	Bohlsee	3.0	0.70	980	1961	4.2	1983	4.1	-0.1	1983(2)	3.5	A, F
58	Feldsee	34.5	9.15	1109	1839-1909(4)	6.7	1983(2)	6.9	ns	1983(3)	6.7	ns
59	Fichtsee	6.5	1.21	598	1916	4.2	1983(2)	4.0	ns	1983	3.9	ns
60	Grosser Arbersee	15.9	7.72	935	1916-49(5)	5.1	1983	4.7	-0.4	1979-83(5)	4.9	A,S,F
61	Kleiner Arbersee	9.7	9.40	918	1900-13(6)	5.1	1983	5.0	-0.1	1982-83(8)	4.4	A, F

Figure 1

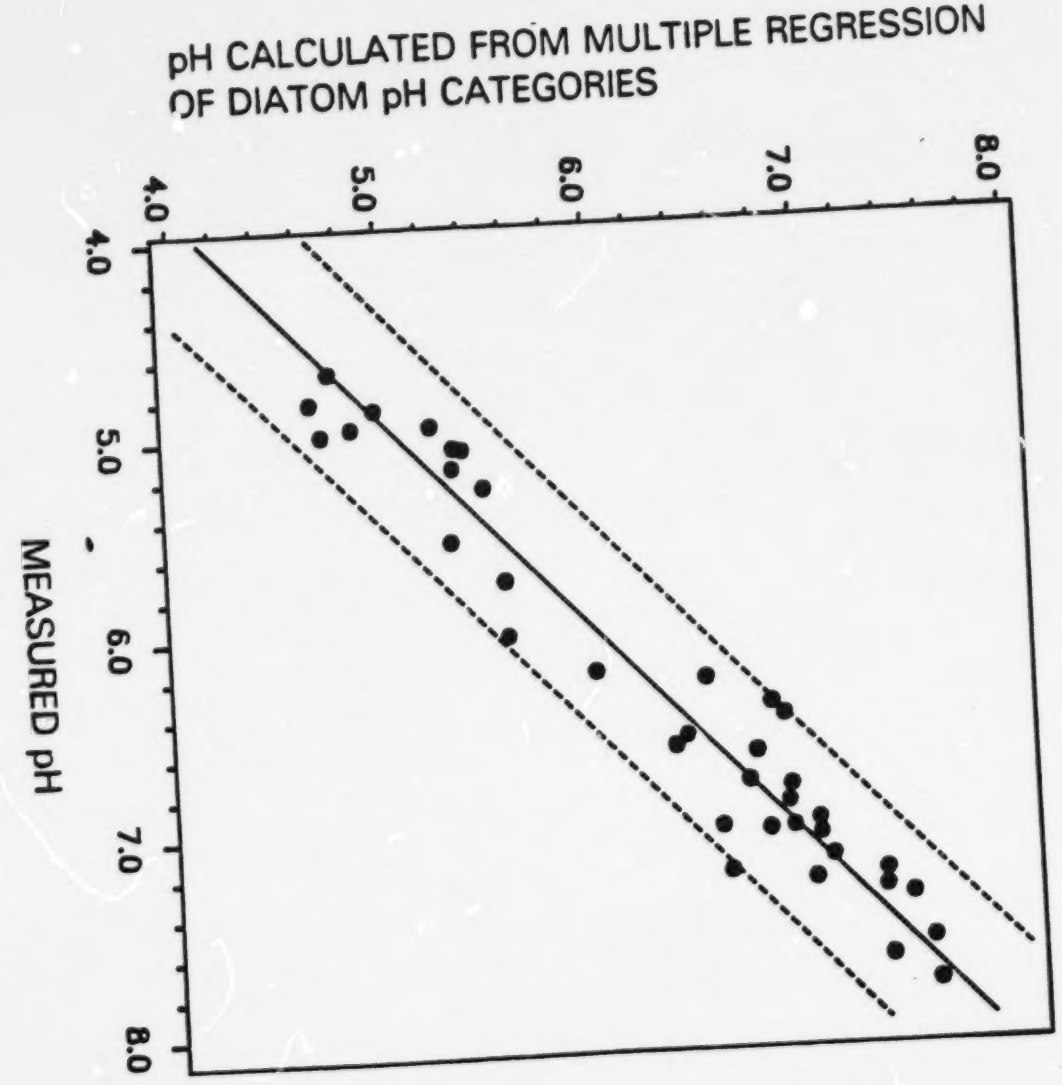

pH CALCULATED FROM MULTIPLE REGRESSION
OF DIATOM pH CATEGORIES
4.0
5.0
6.0
7.0
8.0
MEASURED pH

Figure 2.

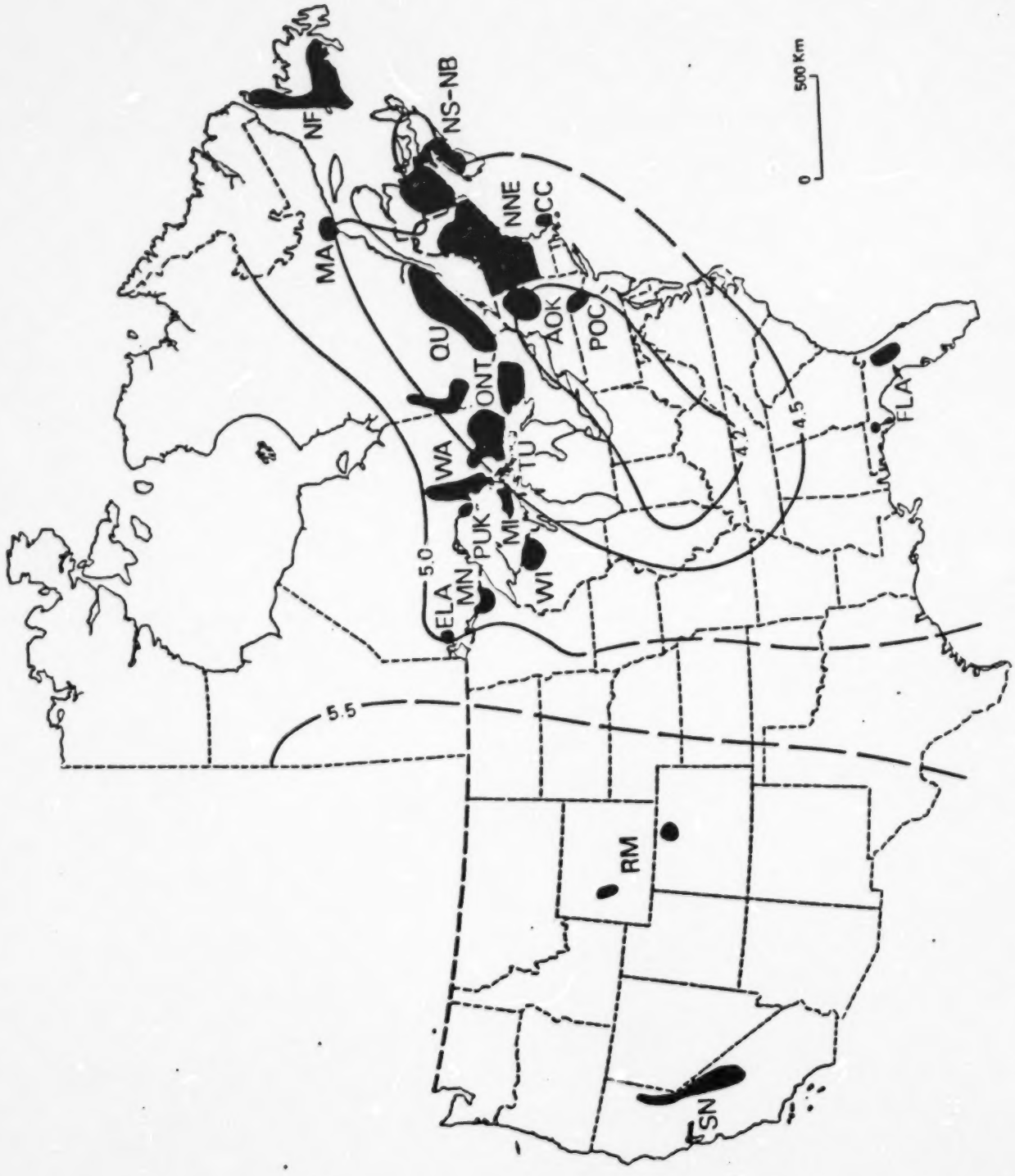

500 Km
0
NF
NS-NB
MA
NNE
CC
QU
AOK
POC
ONT
WA
TU
FLA
4.5
5.0
PUK
MI
MN
WI
ELA
5.5
RM
SN

Figure 3.

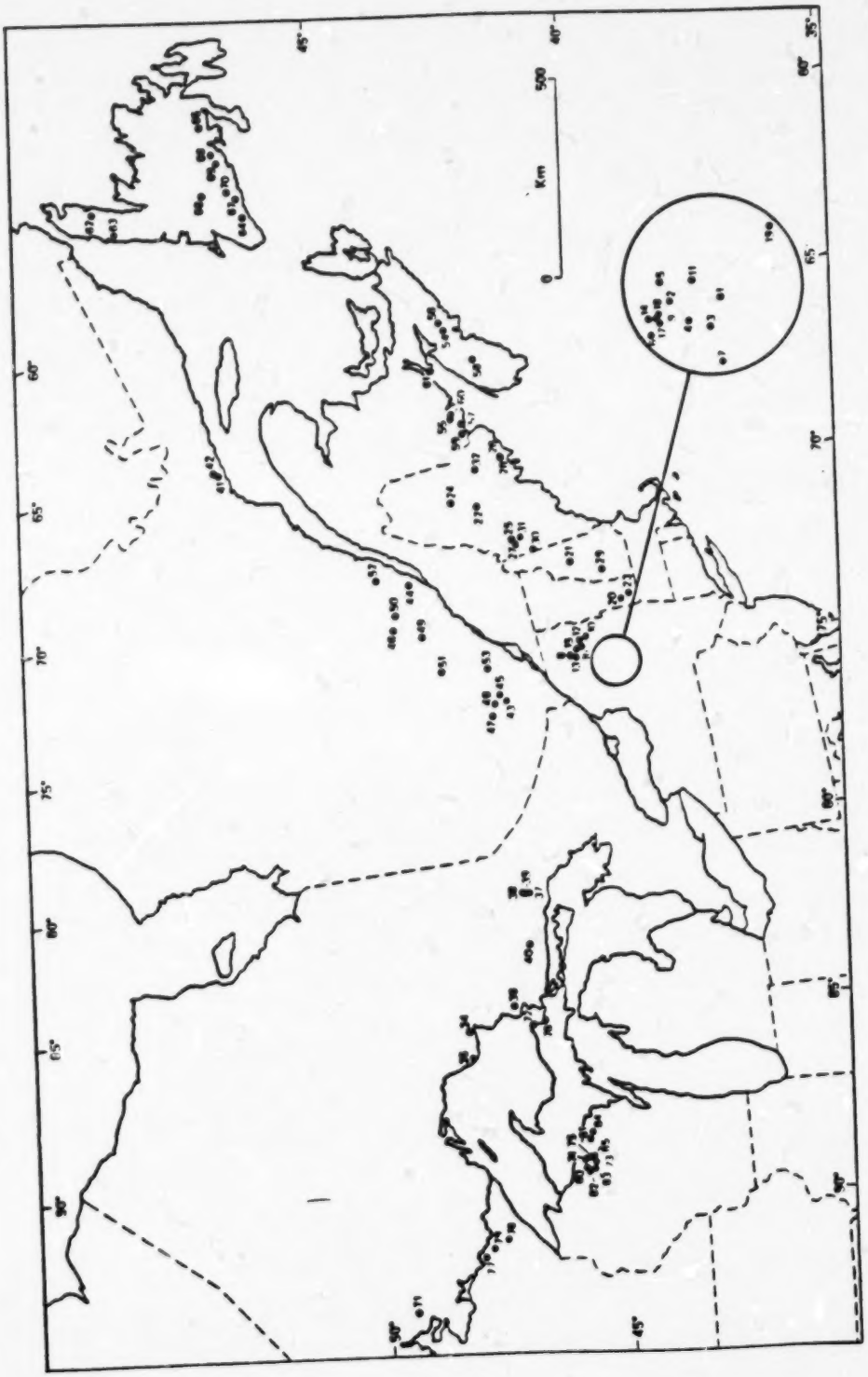


Figure 4.

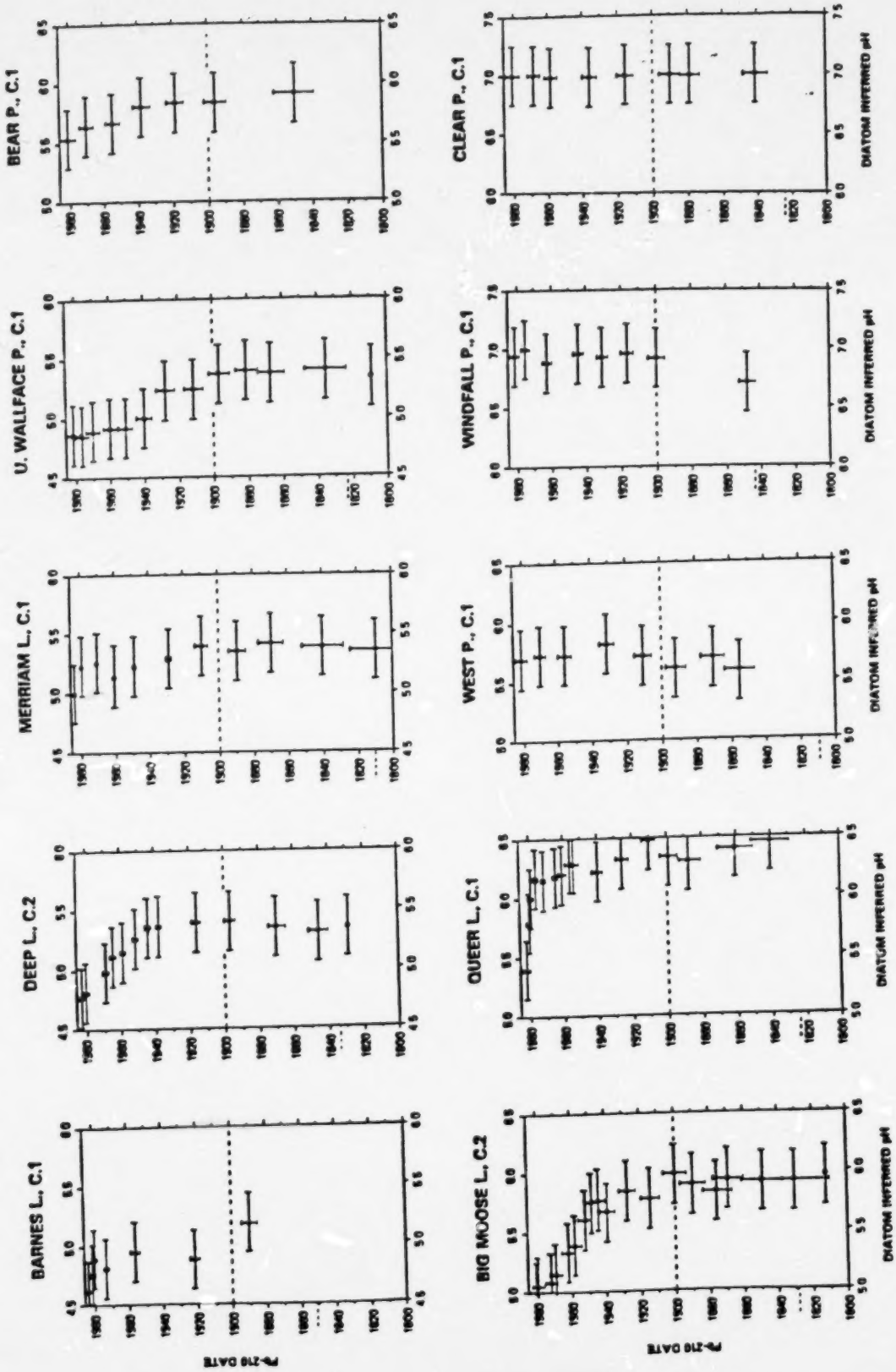

BEAR P., C.1
CLEAR P., C.1
U. WALLFACE P., C.1
WINDFALL P., C.1
MERRIAM L., C.1
WEST P., C.1
DEEP L., C.2
QUEER L., C.1
BARNES L., C.1
BIG MOOSE L., C.2
DIATOM INFERRED pH
Pb-210 DATE

Figure 5

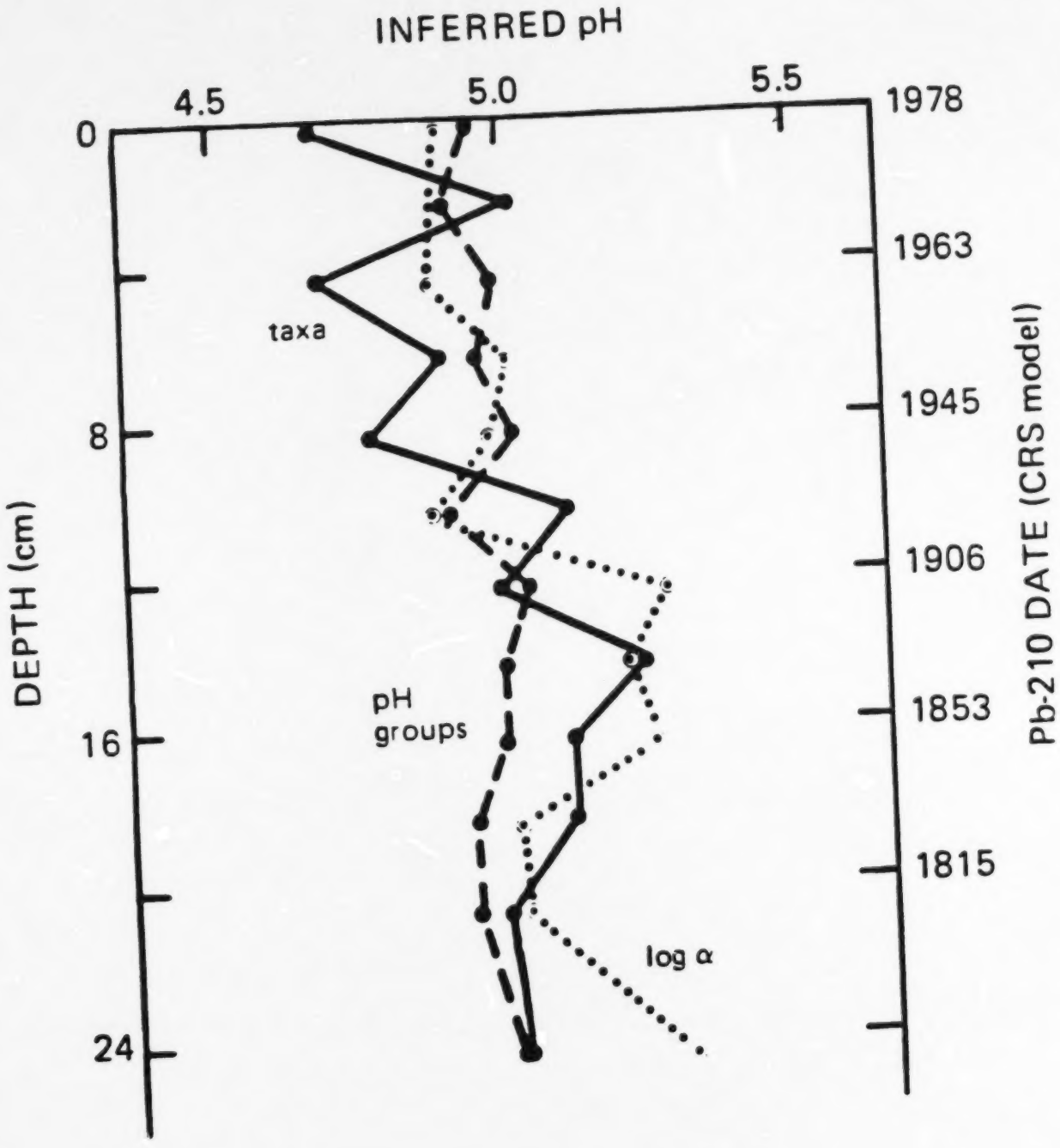

INFERRED pH
4.5
5.0
5.5
0
8
16
24
DEPTH (cm)
1978
1963
1945
1906
1853
1815
Pb-210 DATE (CRS model)
taxa
pH groups
log α

Figure 6.

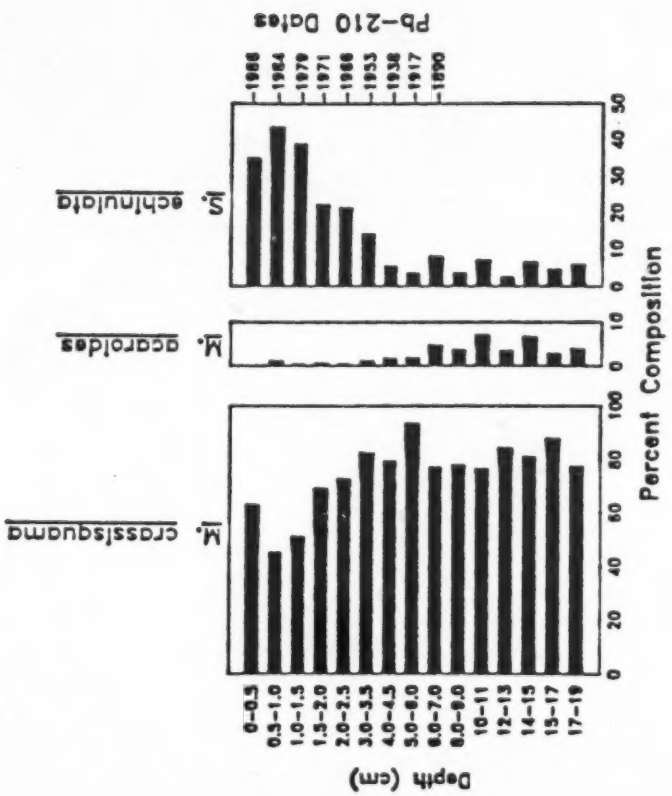

Pb-210 Dates
1988
1984
1979
1971
1966
1953
1938
1917
1890
S. echinulata
M. scarroldes
M. crassisquama
0 10
0 10 20 30 40 50
0 20 40 60 80 100
Percent Composition
0–0.5
0.5–1.0
1.0–1.5
1.5–2.0
2.0–2.5
3.0–3.5
4.0–4.5
5.0–6.0
6.0–7.0
8.0–8.0
10–11
12–13
14–15
15–17
17–19
Depth (cm)

Figure 7.

United Kingdom
pH change 1800 - 1986
• significant change
o no change
0
100 Km

Figure 8.

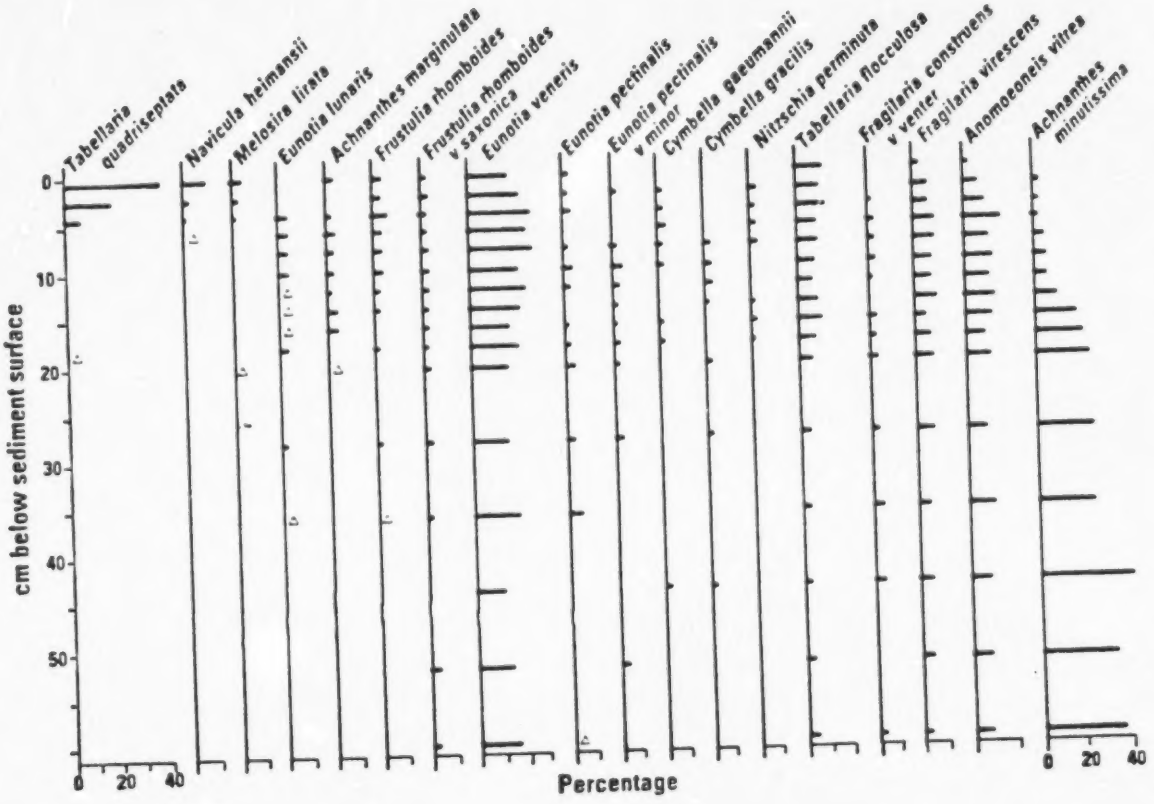

Tabellaria quadriseptata
Navicula heimansii
Melosira lirata
Eunotia lunaris
Achnanthes marginulata
Frustulia rhomboides
Frustulia rhomboides v saxonica
Eunotia veneris
Eunotia pectinalis
Eunotia pectinalis v minor
Cymbella gaeumannii
Cymbella gracilis
Nitzschia perminuta
Tabellaria flocculosa
Fragilaria construens v venter
Fragilaria virescens
Anomoeoneis vitrea
Achnanthes minutissima
cm below sediment surface
0
10
20
30
40
50
0
20
40
Percentage
0
20
40

Figure 9.

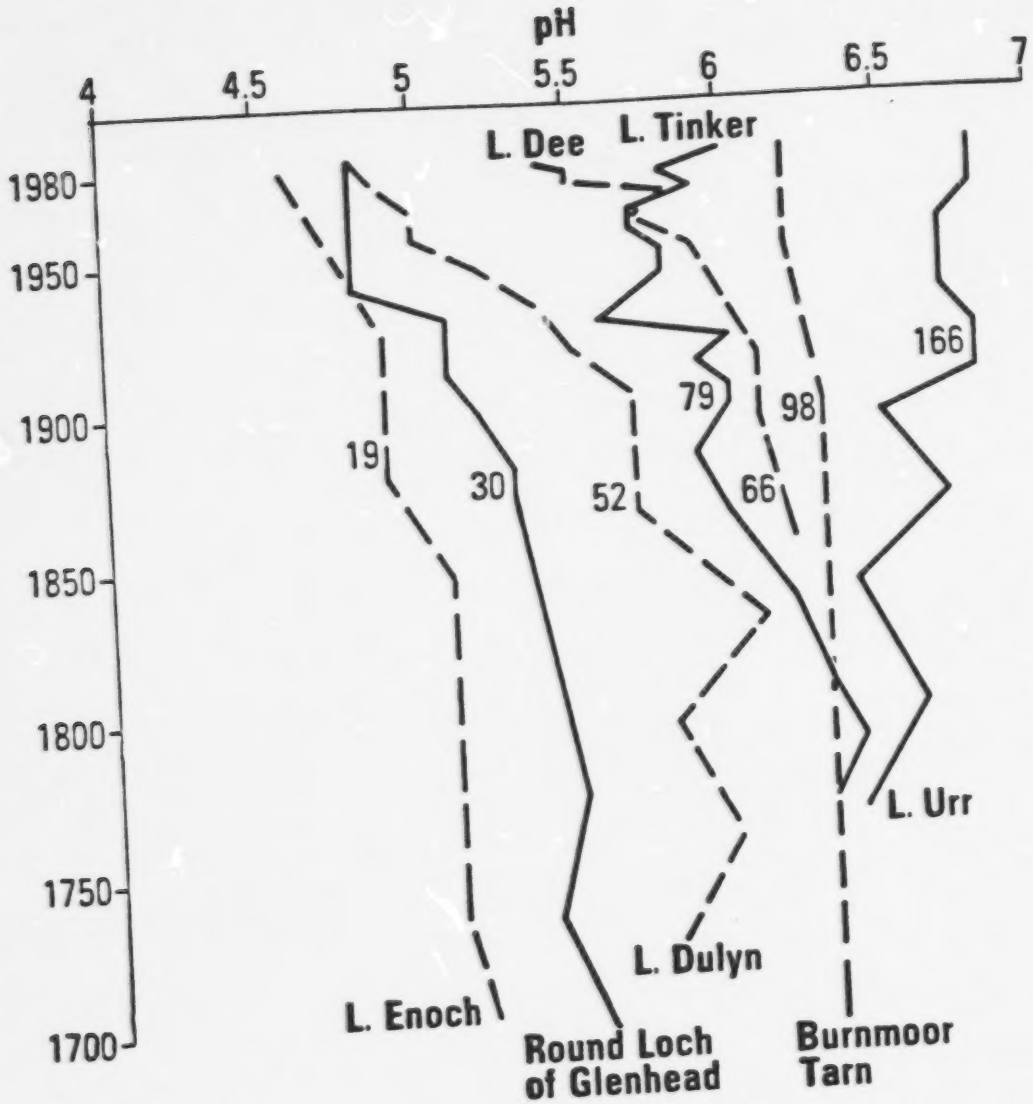

pH
4
4.5
5
5.5
6
6.5
7
1980
1950
1900
1850
1800
1750
1700
L. Dee
L. Tinker
19
30
52
79
98
66
166
L. Urr
L. Dulyn
L. Enoch
Round Loch of Glenhead
Burnmoor Tarn

Figure 10.

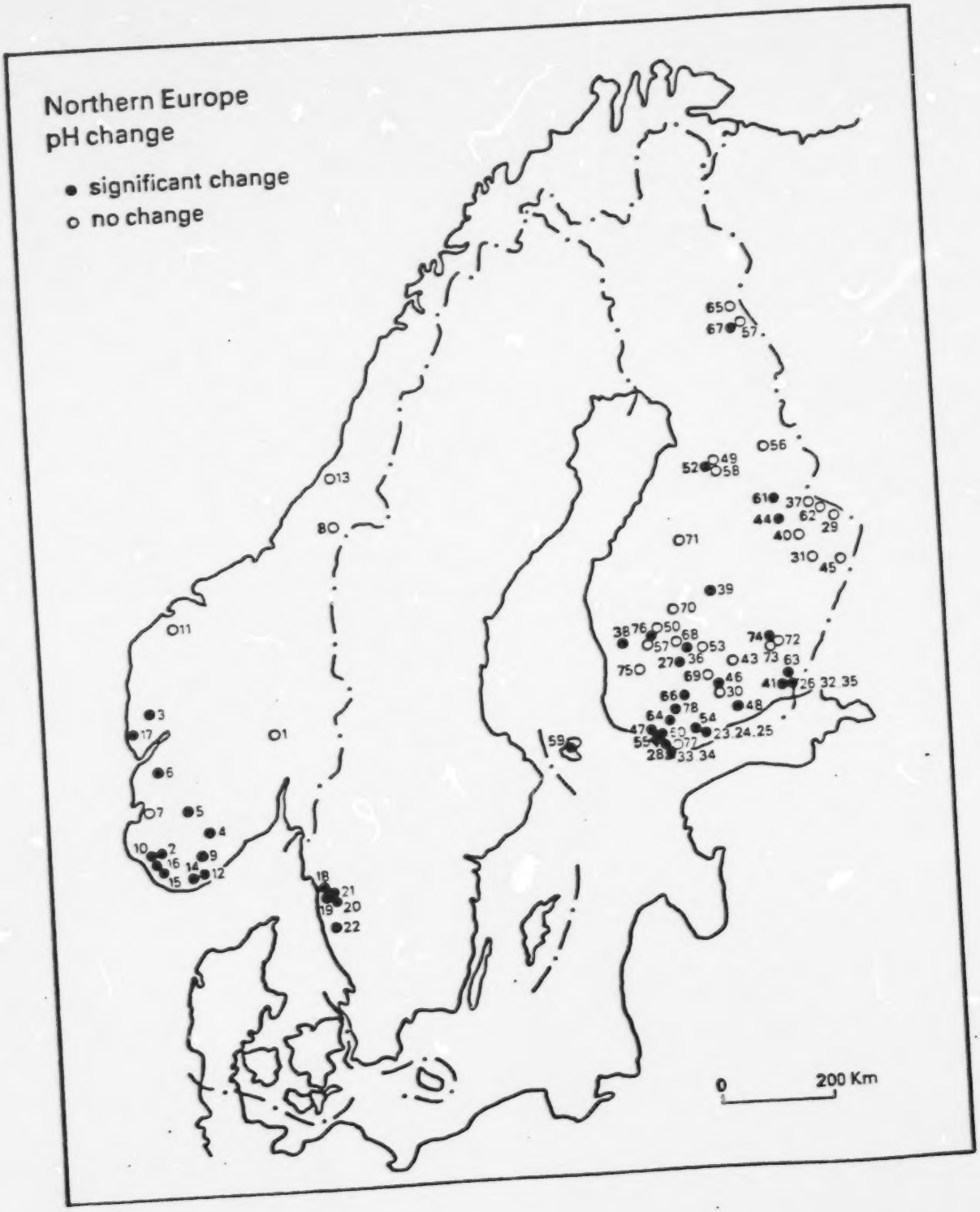

Northern Europe
pH change
• significant change
o no change
0
200 Km

Fig. 11

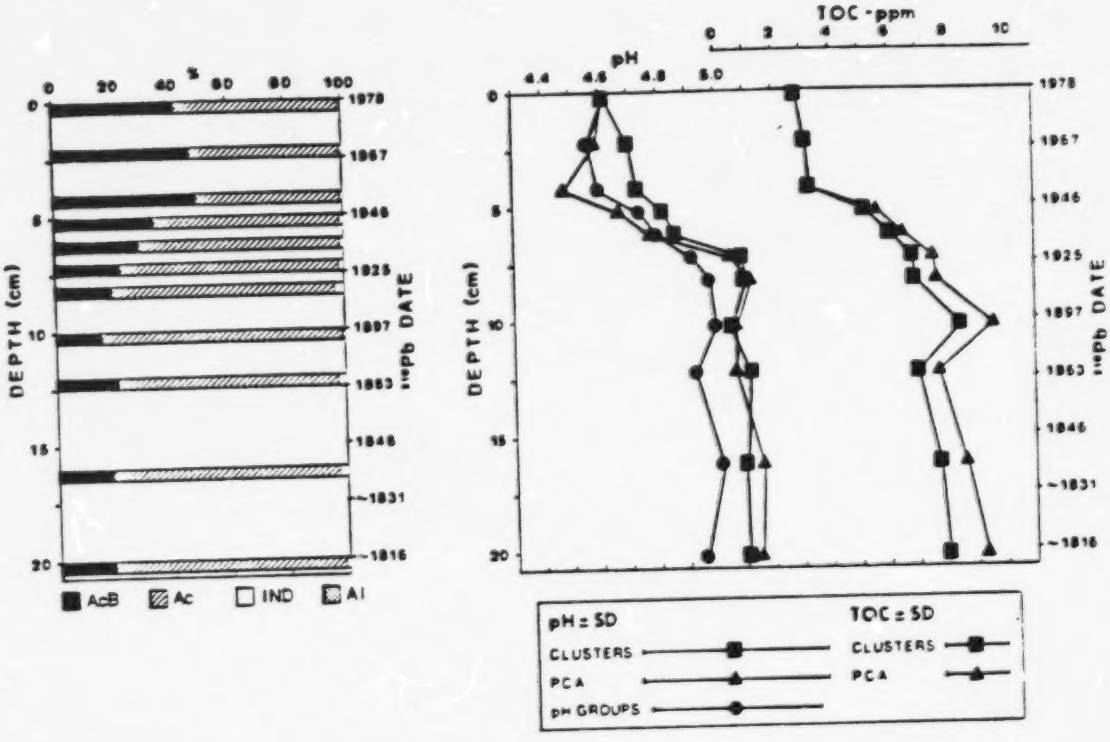

%
0 20 40 60 80 100
DEPTH (cm)
0
5
10
15
20
1978
1967
1946
1925
1897
1883
1848
1831
1816
210Pb DATE
AcB
Ac
IND
AI
TOC - ppm
0 2 4 6 8 10
pH
4.4 4.6 4.8 5.0
pH ± SD
CLUSTERS
PCA
pH GROUPS
TOC ± SD
CLUSTERS
PCA

Fig. 12

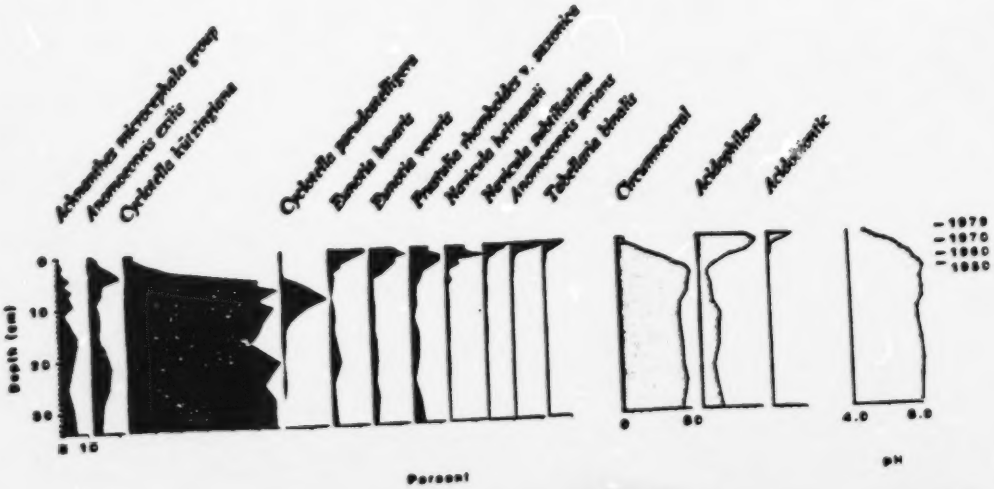

Achnanthes microcephala group
Anomoeoneis exilis
Cyclotella kützingiana
Cyclotella pseudostelligera
Diatoma hiemale
Diatoma vulgare
Fragilaria rhomboides v. austriaca
Navicula krasskei
Navicula subtilissima
Anomoeoneis sphaerophora
Tabellaria binalis
Circumneutral
Acidophilous
Acidobiontic
Depth (cm)
0
10
20
30
1978
1970
1960
1950
0 50
4.0 5.0
Percent
pH

Figure 13.

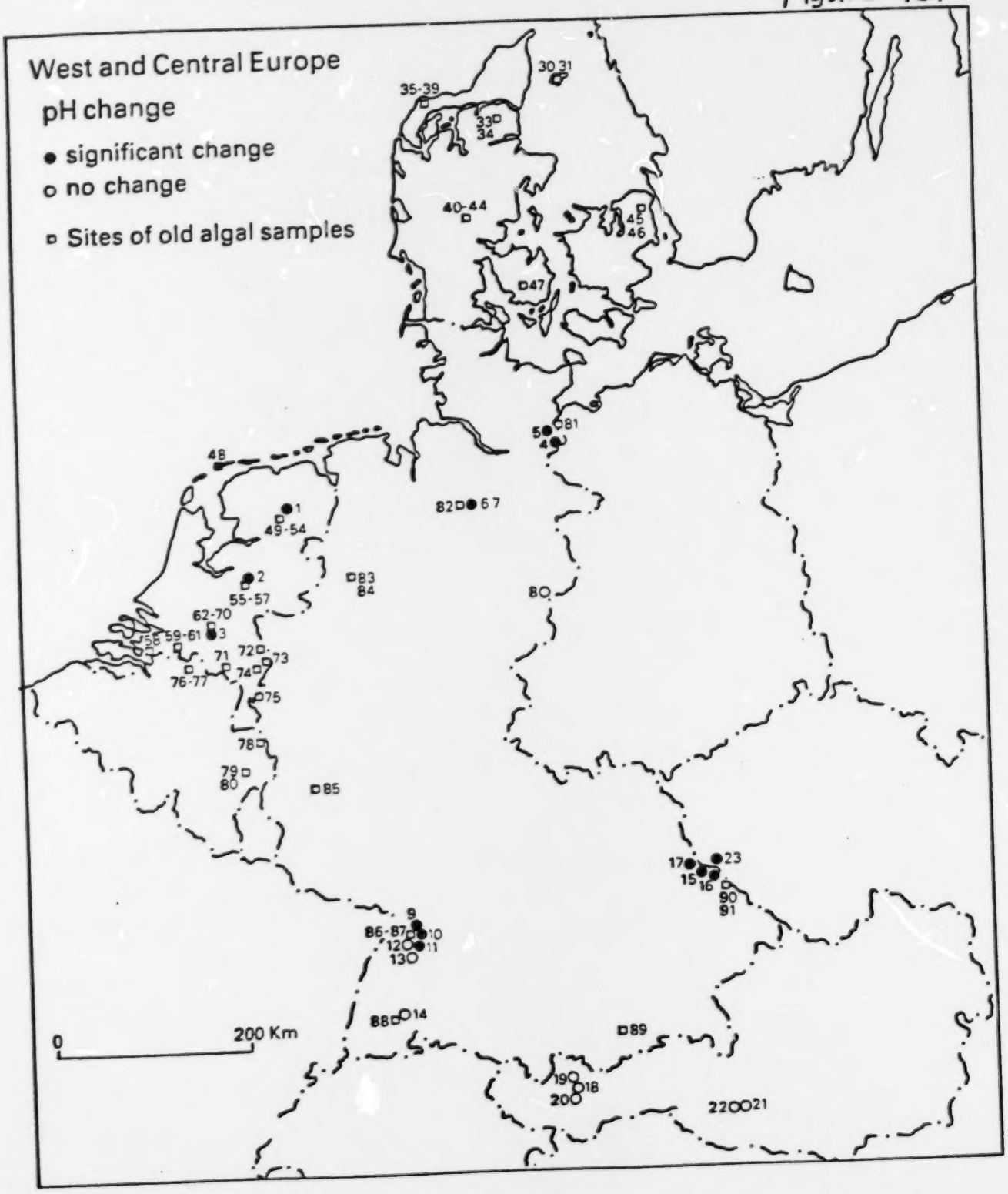
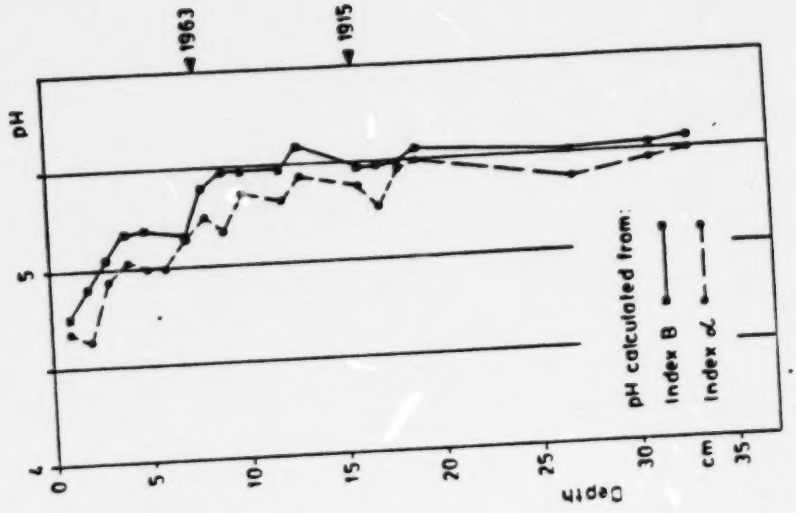

West and Central Europe
pH change
● significant change
○ no change
▫ Sites of old algal samples
0
200 Km

Figure 14.


1963
1915
pH
5
7
0
5
10
15
20
25
30
cm
35
Depth
pH calculated from:
Index B
Index d

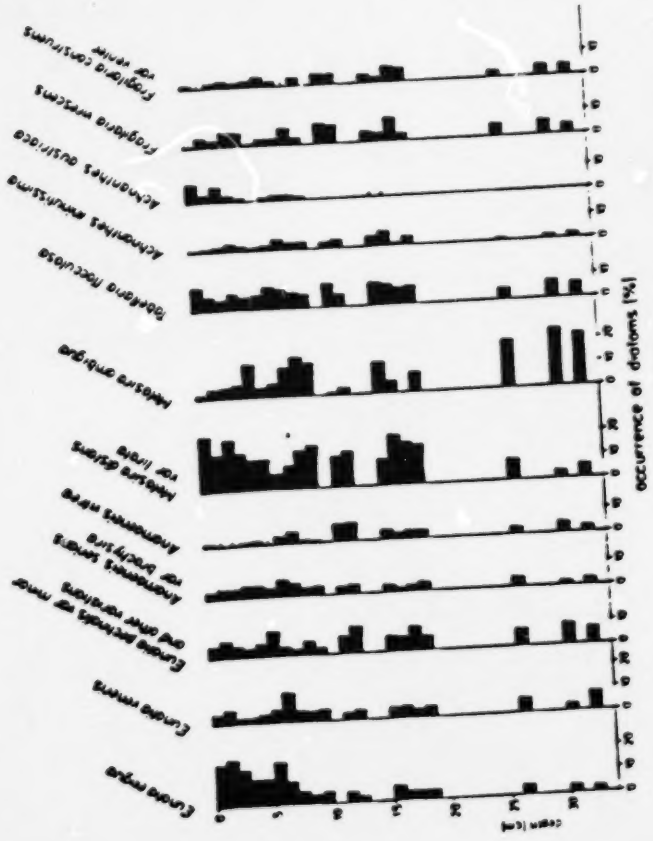
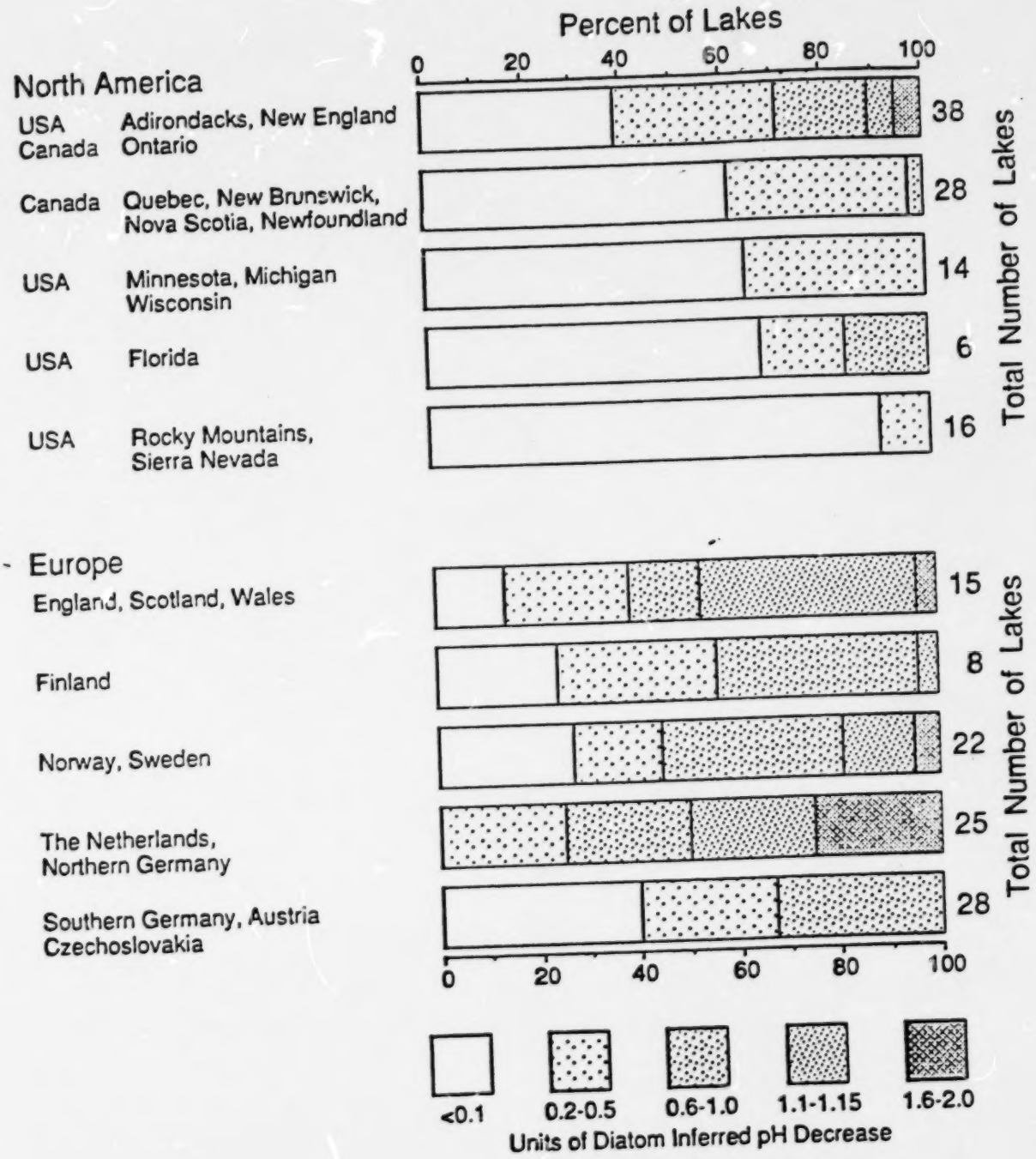

Eunotia incisa
Eunotia minima
Eunotia pectinalis var. minor
and other variations
Anomoeneis sphaera
var. Brachysira
Anomoeneis serians
Melosira distans
var. Lirata
Melosira ambigua
Tabellaria flocculosa
Achnanthes minutissima
Achnanthes austriaca
Frustulia rhomboides
var. saxonica
Frustulia constrictums
var. capitata
occurrence of diatoms (%)
depth (cm)

Figure 15.


Percent of Lakes
0
20
40
60
80
100
North America
USA
Canada
Adirondacks, New England
Ontario
38
Canada
Quebec, New Brunswick,
Nova Scotia, Newfoundland
28
USA
Minnesota, Michigan
Wisconsin
14
USA
Florida
6
USA
Rocky Mountains,
Sierra Nevada
16
Total Number of Lakes
Europe
England, Scotland, Wales
15
Finland
8
Norway, Sweden
22
The Netherlands,
Northern Germany
25
Southern Germany, Austria
Czechoslovakia
28
Total Number of Lakes
0
20
40
60
80
100
<0.1
0.2-0.5
0.6-1.0
1.1-1.15
1.6-2.0
Units of Diatom Inferred pH Decrease